THE PROCEEDINGS OF
THE PHYSICAL SOCIETY

Section A

VOL. 65, PART 7 1 July 1952 No. 391A

The Solution of the Fluctuation Problem in Nucleon Cascade Theory: Homogeneous Nuclear Matter

By H. MESSEL
Department of Mathematical Physics, University of Adelaide, Australia

MS. received 2nd November 1951

ABSTRACT. Solutions are presented for the general number distribution function in cascade theory and for the *k*th moments of the distribution. The Jánossy *G*-equation is solved. The methods of solution developed are applicable to the similar problem in the electron–photon case.

§ 1. INTRODUCTION

The fluctuation problem for nucleonic showers is of the same complexity as that found in electron–photon shower theory. In both cases one is eventually faced with the task of solving one or more partial integro-differential equations. Jánossy (1949) in an effort to solve the fluctuation problem was successful in setting up the fundamental equation for a special case of the general number distribution function. His so-called *G*-equation was of great value, for from it one could derive the partial integro-differential equations for higher moments of the distribution. Recently (Messel 1951 a, Jánossy and Messel 1951) the second moments of the distribution were found both for homogeneous nuclear matter and a finite absorber. However, little headway was made in solving for the distribution function itself or for its moments higher than the second.

In this paper we shall give the solutions for the general number distribution function in homogeneous nuclear matter and a number of subsidiary functions arising therefrom. Furthermore, we shall present solutions for the *n*th moments as well as a formal solution of the Jánossy *G*-equation.

Throughout the work which follows we shall make use of one assumption only with respect to the cross section for nucleon–nucleon collisions, e.g. the cross section for the production of secondary and recoil nucleons of energy E_1 and E_2 by a primary nucleon of energy E_0 is a homogeneous function of the primary and secondary energies, thus :

$$w(E_0; E_1, E_2)\, dE_1\, dE_2 = w(E_1/E_0, E_2/E_0)E_0^{-2}\, dE_1\, dE_2. \qquad \ldots\ldots(1)$$

Otherwise the function w is perfectly general.

§ 2. THE DIFFUSION EQUATION FOR THE NUMBER DISTRIBUTION FUNCTION AND ITS SOLUTION

The most general distribution function in nucleon cascade theory is denoted by $\phi_n(\epsilon_1, \ldots \epsilon_n; x)d\epsilon_1 \ldots d\epsilon_n$. It defines the differential probability of finding the n nucleons with energies ϵ_i, $\epsilon_i + d\epsilon_i (\epsilon_i = E_i/E_0)$ in the order $\epsilon_1, \ldots, \epsilon_n$ at the depth x in homogeneous nuclear matter, due to a single incident primary nucleon of energy E_0. From the function ϕ_n all other distribution functions of importance in the theory may be obtained. The function

$$F_n(\epsilon_1, \ldots, \epsilon_n; x) = \sum_{\text{perm}} \phi_n(\epsilon_1, \ldots, \epsilon_n; x) = n\,!\,\phi_n(\epsilon_1, \ldots, \epsilon_n; x) \quad \ldots\ldots(2)$$

expresses the probability of finding any n particles with energies ϵ_i, $\epsilon_i + d\epsilon_i$ at the depth x. Its equation has been given previously in a number of papers (cf. Messel 1951 b)

$$\frac{\partial}{\partial x} F_n(\epsilon_1, \ldots, \epsilon_n; x) = -nF_n(\epsilon_1, \ldots, \epsilon_n; x)$$
$$+ \sum_{k \neq l} \int_0^\infty F_{n-1}(\epsilon_1', \ldots, \epsilon_{n-2}', \epsilon; x) w(\epsilon_k/\epsilon, \epsilon_l/\epsilon)\epsilon^{-2}\, d\epsilon, \quad \ldots\ldots(3)$$

where the primes signify that neither ϵ_k nor ϵ_l are included among the $n-2$ energy ratios ϵ_i'. It may be shown (Messel 1951 b) that the dependence on the depth x of the F_n may be split off, thus

$$F_n(\epsilon_1, \ldots, \epsilon_n; x) = f_n(x) A_n(\epsilon_1, \ldots, \epsilon_n) \quad \ldots\ldots(4)$$

where

$$f_n(x) = \frac{1}{n!} \int_0^\infty \ldots \int_0^\infty F_n(\epsilon_1, \ldots, \epsilon_n; x)\, d\epsilon_1 \ldots d\epsilon_n = \mathrm{e}^{-x}(1 - \mathrm{e}^{-x})^{n-1}. \quad \ldots\ldots(5)$$

The function $f_n(x)$ expresses the probability of finding n particles of arbitrary energies at the depth x. Also $\Sigma_n f_n(x) = 1$. From (3) and (4) it is seen that

$$A_n(\epsilon_1, \ldots, \epsilon_n) = \frac{1}{n-1} \sum_{k \neq l} \int_0^\infty A_{n-1}(\epsilon_1', \ldots, \epsilon_{n-2}', \epsilon) w(\epsilon_k/\epsilon, \epsilon_l/\epsilon)\epsilon^{-2}\, d\epsilon. \quad \ldots\ldots(6)$$

Next we define the n-fold Mellin transform of A_n as

$$M_n(s_1, \ldots, s_n) = \int_0^\infty \ldots \int_0^\infty \epsilon_1^{s_1} \ldots \epsilon_n^{s_n} A_n(\epsilon_1, \ldots, \epsilon_n)\, d\epsilon_1 \ldots d\epsilon_n. \quad \ldots\ldots(7)$$

Apply the n-fold transform to (6) and find

$$M_n(s_1, \ldots, s_n) = \frac{1}{n-1} \sum_{k \neq l} W(s_k, s_l) M_{n-1}(s_1', \ldots, s_{n-2}', s_k + s_l), \quad \ldots\ldots(8)$$

where $$W(s_k, s_l) = \int_0^\infty \int_0^\infty \epsilon_k^{s_k} \epsilon_l^{s_l} w(\epsilon_k, \epsilon_l)\, d\epsilon_k\, d\epsilon_l. \quad \ldots\ldots(9)$$

Our initial condition is determined, since there can only be one nucleon of energy E_0 at the depth $x = 0$, thus $A_1(\epsilon) = \delta(1 - \epsilon)$ and $M_1(s) = 1$. The solution of (8) is now immediate and may be given as

$$M_n(s_1, \ldots, s_n) = (2^{n-1}/(n-1)\,!)\Lambda_n(s_1, \ldots, s_n), \quad \ldots\ldots(10)$$

where*

$$\Lambda_n(s_1, \ldots, s_n) = \sum_{a \neq b}{}' W(s_a, s_b) \sum_{a \neq b \neq c \neq d}^{2} W(s_c, s_d) \ldots \sum_{a \neq b \neq \ldots \neq y \neq z}^{n-1} W(s_y, s_z). \quad \ldots(11)$$

---

* Equation (11) may be given in the compact form

$$\Lambda_n(s_1, \ldots, s_n) = \prod_{v=n-1}^{1} \Sigma' W(s_v, s_{v+1} + \ldots, s_n),$$

where Σ' signifies summation over all combinations of the $v+1$ variables $s_1, \ldots, s_v, s_{v+1} + \ldots, s_n$, taken two at a time.

The factor 2^{n-1} in (10) arises due to the symmetry of the W function. The total number of terms in (11) arising from the $(n-1)$-fold summation is $n!\,(n-1)!/2^{n-1}$. A few words should be added regarding the above summations. The superscripts beside each summation refer to the order in which it is most convenient to carry out the summation. The first summation simply yields W functions for all possible combinations of the n variables $s_i, i=1, \ldots, n$ taken two at a time. The second summation then gives W functions for all possible combinations of the $n-1$ variables $s_a+s_b, s_1', \ldots, s_{n-2}'$ taken two at a time, similarly the third summation gives W functions for all possible combinations of the $n-2$ variables $s_c+s_d, s_1'', \ldots, s_{n-3}''$ taken two at a time, and so on for the remainder of the summations. We give in the Appendix the function M_n found using (11) for the cases $n=2$ and 4. Using (5), (10), (11) and applying an n-fold inverse Mellin transform we find for the solution of the diffusion eqn. (3),

$$F_n(\epsilon_1, \ldots, \epsilon_n; x) = (2^{n-1}/(n-1)!)f_n(x)L_n\Lambda_n(s_1, \ldots, s_n), \quad \ldots\ldots(12)$$

where

$$L_n = \frac{1}{(2\pi i)^n}\int_{u_1-i\infty}^{u_1+i\infty} \cdots \int_{u_n-i\infty}^{u_n+i\infty} \epsilon_1^{-(s_1+1)} \ldots \epsilon_n^{-(s_n+1)}\, ds_1 \ldots ds_n, \quad \ldots\ldots(13)$$

is the inverse Mellin transform operator, operating on the integrand Λ_n in (12).

From (12) it is a straightforward procedure to obtain the formal expressions for other distribution functions of interest in the theory. For instance, the probability $\Phi(\epsilon, n_1, n_2; x)$ of finding n_1 nucleons with energies greater than ϵE_0 and n_2 particles with energies less than ϵE_0 is obtained from (12) by integrating over n_1 of the variables $\epsilon_i, i=1, \ldots, n$ from ϵ to 1 and over the remaining n_2 variables from 0 to ϵ. Thus

$$\Phi(\epsilon, n_1, n_2; x) =$$
$$(2^{n-1}/n_1!\,n_2!\,(n-1)!)f_n(x)\int_0^\epsilon \underset{n_2}{\cdots}\int_0^\epsilon\int_\epsilon^1 \underset{n_1}{\cdots}\int_\epsilon^1 L_n\Lambda_n(s_1, \ldots, s_n)\, d\epsilon_1 \ldots d\epsilon_n. \quad (14)$$

So far we have been concerned with homogeneous nuclear matter only. However, on the assumption that nuclei are spherical in shape, we may apply our results directly to nuclei by simply replacing the factor $f_n(x)$ in the above expressions by $R_n(D_A)$ where

$$R_n(D_A) = \int_0^1 2xf_n(D_Ax)\,dx \quad \ldots\ldots(15)$$

and D_A is the average number of collisions suffered by a nucleon in making a diametrical passage through a nucleus of atomic weight A.

§ 3. THE DIFFUSION EQUATION AND SOLUTION FOR $C_n(\epsilon_1, \ldots, \epsilon_n; x)$

Let $C_n(\epsilon_1, \ldots, \epsilon_n; x)d\epsilon_1 \ldots d\epsilon_n$ be the differential probability of finding one particle in each of the energy intervals $\epsilon_i, \epsilon_i+d\epsilon_i$ and any number of particles with arbitrary energies at a depth x in homogeneous nuclear matter, due to a primary nucleon of unit energy. The n-fold Mellin transform of C_n is given by

$$V_n(s_1, \ldots, s_n; x) = \int_0^\infty \cdots \int_0^\infty \epsilon_1^{s_1} \ldots \epsilon_n^{s_n} C_n(\epsilon_1, \ldots, \epsilon_n; x)\, d\epsilon_1 \ldots d\epsilon_n. \quad \ldots(16)$$

The relation between F_n and C_n is expressed by

$$C_n(\epsilon_1, \ldots, \epsilon_n; x) = \sum_{v=0}^{\infty} \frac{1}{v!} \int_0^\infty \cdots \int_0^\infty F_{n+v}(\epsilon_1, \ldots, \epsilon_{n+v}; x)\, d\epsilon_{n+1} \ldots d\epsilon_{n+v}. \qquad \ldots\ldots(17)$$

Since the function F_n has been determined in (12), the solution for C_n is given by (17). However, owing to the complicated nature of the result for F_n it is difficult to obtain directly from (17) explicit values for C_n for different values of n.

Explicit results for V_n are required for finding the nth moments of the number distribution function as well as in all problems dealing with the development of the cascade in a finite absorber. In previous work (Jánossy and Messel 1951) we developed a method for obtaining V_n without knowing the solution of the diffusion equation for F_n. The method used was far from straightforward, and it would have been difficult to obtain explicit results for values of n equal to or greater than 3. We now present a new method for obtaining V_n. The diffusion equation for C_n is given and its solution obtained in the form of a simple recurrence relation. One may proceed in either of two ways to obtain the diffusion equation. The first method is to use (17) and the diffusion equation for F_n; the second is to set up the equation directly.*

Consider the n particles in the energy intervals $\epsilon_i, \epsilon_i + d\epsilon_i$, $i = 1, \ldots, n$, at the depth x. At some previous depth $x - \theta$ the last change in number must have taken place in such a manner that we obtain the n particles in the energy intervals $d\epsilon_i$. There exist only two possibilities:

(*a*) At the depth $x-\theta$, $n-1$ of the particles may have attained their final energies, then one of the particles undergoes a collision giving rise to the nth particle. Once this collision has occurred no further change may take place. The probability for this process is given by

$$\sum_{l=1}^{n} \mathrm{e}^{-n\theta}\, d\theta \int_0^\infty C_n(\epsilon_1', \ldots, \epsilon_{n-1}', \epsilon; x-\theta) W(\epsilon_l/\epsilon)\epsilon^{-1}\, d\epsilon$$

with

$$W(\epsilon) = \int_0^\infty \{w(\epsilon, \epsilon_1) + w(\epsilon_1, \epsilon)\}\, d\epsilon_1 = \int_0^\infty \overline{w}(\epsilon, \epsilon_1)\, d\epsilon_1 = 2\int_0^\infty w(\epsilon, \epsilon_1)\, d\epsilon_1, \qquad \ldots\ldots(18)$$

because of the symmetry of the cross section $w(\epsilon_1, \epsilon_2)$.

(*b*) Similarly for the second case where we have $n-2$ particles in their final energy intervals at the depth $x-\theta$. One of the particles undergoes a collision giving rise to the $(n-1)$th and nth particles. The probability for this process is

$$\sum_{J \neq i} \mathrm{e}^{-n\theta}\, d\theta \int_0^\infty C_{n-1}(\epsilon_1', \ldots, \epsilon_{n-2}', \epsilon; x-\theta) w(\epsilon_J/\epsilon, \epsilon_l/\epsilon)\epsilon^{-2}\, d\epsilon.$$

Thus we find

$$C_n(\epsilon_1, \ldots, \epsilon_n; x) = \int_0^x \mathrm{e}^{-n\theta} \left\{ \int_0^\infty \sum_{l=1}^{n} C_n(\epsilon_1', \ldots, \epsilon_{n-1}', \epsilon; x-\theta) W(\epsilon_l/\epsilon)\epsilon^{-1}\, d\epsilon \right.$$
$$\left. + \int_0^\infty \sum_{J \neq l} C_{n-1}(\epsilon_1', \ldots, \epsilon_{n-2}', \epsilon; x-\theta) w(\epsilon_J/\epsilon, \epsilon_l/\epsilon)\epsilon^{-2}\, d\epsilon \right\} d\theta. \qquad \ldots\ldots(19)$$

---

* We believe Jánossy gave in an unpublished manuscript the same equation as we give. He was not successful, however, in obtaining its solution.

This may be transformed by differentiation into the form

$$\frac{\partial}{\partial x} C_n(\epsilon_1, \ldots, \epsilon_n; x) = -nC_n(\epsilon_1, \ldots, \epsilon_n; x)$$

$$+ \int_0^\infty \sum_{l=1}^{n} C_n(\epsilon_1', \ldots, \epsilon_{n-1}', \epsilon; x) W(\epsilon_l/\epsilon)\epsilon^{-1}\, d\epsilon$$

$$+ \int_0^\infty \sum_{J \neq l} C_{n-1}(\epsilon_1', \ldots, \epsilon_{n-2}', \epsilon; x) w(\epsilon_J/\epsilon, \epsilon_l/\epsilon)\epsilon^{-2}\, d\epsilon. \quad \ldots\ldots(20)$$

To solve (20), take the n-fold Mellin transform defined by (16) and find

$$\frac{\partial}{\partial x} V_n(s_1, \ldots, s_n; x) = -nV_n(s_1, \ldots, s_n; x) + \sum_{l=1}^{n} W^*(s_l) V_n(s_1, \ldots, s_n; x)$$

$$+ 2 \sum_{J \neq l} W(s_J, s_l) V_{n-1}(s_1', \ldots, s_{n-2}', s_J + s_l; x), \quad \ldots\ldots(21)$$

where

$$W^*(s) = \int_0^\infty \epsilon^s W(\epsilon)\, d\epsilon. \quad \ldots\ldots(22)$$

If we write $\alpha(s) = 1 - W^*(s)$ then the solution of (21) becomes

$$V_n(s_1, \ldots, s_n; x) = 2 \exp\left\{-x \sum_{i=1}^{n} \alpha(s_i)\right\} \int_0^x \exp\left\{\theta \sum_{i=1}^{n} \alpha(s_i)\right\} \sum_{J \neq l} W(s_J, s_l)$$

$$\times V_{n-1}(s_1', \ldots, s_{n-2}', s_J + s_l; \theta)\, d\theta. \quad \ldots\ldots(23)$$

From our initial condition we obtain for the case $n = 1$, $V_1(s; x) = \exp\{-\alpha(s)x\}$. Explicit results for higher values of n may now be obtained by using (23). The integration though lengthy for high values of n is elementary. We give in the Appendix the expressions for V_2 and V_3. The general expression for V_n can also be given but because of its length we omit it here.†

Applying an inverse n-fold Mellin transform to (23) we find for our solution of C_n

$$C_n(\epsilon_1, \ldots, \epsilon_n; x) = L_n V_n(s_1, \ldots, s_n; x), \quad \ldots\ldots(24)$$

which is necessarily equivalent to the solution given by (17). The above result may be extended to nuclei by applying the averaging operator $\int_0^{D_A} 2x\, dx/D_A^2$ to it.

§ 4. THE *G*-EQUATION, ITS SOLUTION AND ALTERNATIVE EQUATIONS FOR $F_n(\epsilon_1, \ldots, \epsilon_n; x)$ AND $C_n(\epsilon_1, \ldots, \epsilon_n; x)$

Jánossy (1949) set up the following diffusion equation for $\Phi(\epsilon, n_1, n_2; x)$ defined by us in § 2,

$$\Phi(\epsilon, n_1, n_2; x) = \int_0^x e^{-(x-\theta)}\, d\theta \sum_{\substack{n_1'+n_1''=n_1 \\ n_2'+n_2''=n_2}} \int_0^\infty \int_0^\infty \Phi\left(\frac{\epsilon}{\epsilon_1}, n_1', n_2'; \theta\right)$$

$$\times \Phi\left(\frac{\epsilon}{\epsilon_2}, n_1'', n_2''; \theta\right) w(\epsilon_1, \epsilon_2)\, d\epsilon_1\, d\epsilon_2. \quad \ldots\ldots(25)$$

---

† The explicit expression for C_n and V_n is given in full by Messel and Potts (1952): see their eqn. (30), where the recurrence relation for V_n is completely solved.

In a similar manner we have set up alternative diffusion equations for F_n and C_n which are practically identical in form with that of (25).

$$F_n(E_0;E_1,\ldots,E_n;x)=\int_0^x e^{-(x-\theta)}\,d\theta\sum_{n_1+n_2=n}\int_0^\infty\int_0^\infty F_{n_1}(E_0';E_1',\ldots,E_{n_1}';\theta)$$
$$\times F_{n_2}(E_0'',E_1'',\ldots,E_{n_2}'';\theta)w(E_0'/E_0,E_0''/E_0)E_0^{-2}\,dE_0'\,dE_0'',\qquad\ldots\ldots(26)$$

where the summation is carried out over all possible ways of obtaining $n_1+n_2=n$. Note that whenever either n_1 or $n_2=0$ then the product $F_{n_1}F_{n_2}$ necessarily vanishes. The primes on the E_i indicate that none of the E_i' are included among the E_i''.

$$C_n(E_0;E_1,\ldots,E_n;x)=\int_0^x e^{-(x-\theta)}\,d\theta\sum_{n_1+n_2=n}\int_0^\infty\int_0^\infty C_{n_1}(E_0';E_1',\ldots,E_{n_1}';\theta)$$
$$\times C_{n_2}(E_0'';E_1'',\ldots:E_{n_2}'';\theta)w(E_0'/E_0,E_0''/E_0)E_0^{-2}\,dE_0'\,dE_0'',\qquad\ldots\ldots(27)$$

where the summation is carried out over all possible ways of obtaining $n_1+n_2=n$. In this instance when either n_1 or $n_2=0$ the product $C_{n_1}C_{n_2}$ does *not* vanish.

It is a peculiar circumstance that both eqns. (26) and (27) may be solved *directly* by means of a n-fold Mellin transform and solutions obtained which may be shown to be equivalent to those already given by us in (12), (17) and (23) respectively, but not eqn. (25), even though it is only a special case of (26). The solution of (25) for $\Phi(\epsilon,n_1,n_2;x)$ is, of course, given by (14), but this was obtained by solving for the general probability distribution function F_n first.

The difficulty of solving (25) for $\Phi(\epsilon,n_1,n_2;x)$ led Jánossy to introduce a generating function defined by

$$G(\epsilon,U;x)=\sum_N U^N\Phi(\epsilon,N;x),\qquad\ldots\ldots(28)$$

in which U is written for u_1 and u_2, N for n_1, n_2 and U^N for $u_1^{n_1}u_2^{n_2}$. Equation (25) could then be put in the seemingly simplified form

$$\frac{\partial}{\partial x}G(\epsilon,U;x)+G(\epsilon,U;x)=\int_0^\infty\int_0^\infty G\left(\frac{\epsilon}{\epsilon_1},U;x\right)G\left(\frac{\epsilon}{\epsilon_2},U;x\right)w(\epsilon_1,\epsilon_2)\,d\epsilon_1\,d\epsilon_2\qquad\ldots\ldots(29)$$

with the initial condition
$$G(\epsilon,u_1,u_2;0)=\begin{cases}u_1 & \text{for}\quad \epsilon<1,\\ u_2 & \text{for}\quad \epsilon>1.\end{cases}\qquad\ldots\ldots(30)$$

The solution of the G-equation is of no interest in itself; however, for the sake of completeness we give its solution below:

$$G(\epsilon,U;x)=\sum_N\frac{1}{n_1!\,n_2!}f_n(x)U^N\int_0^\epsilon\underset{n_2}{\cdots}\int_0^\epsilon\int_\epsilon^1\underset{n_1}{\cdots}\int_\epsilon^1 L_nM_n(s_1,\ldots,s_n)\,d\epsilon_1\ldots d\epsilon_n\qquad\ldots\ldots(31)$$

obtained from (10) and (28).

§ 5. THE *k*TH MOMENTS OF THE DISTRIBUTION FUNCTION

The kth moment of the distribution function giving the probability of finding n particles with energies greater than ϵE_0 and an arbitrary number of particles with energies less than ϵE_0 at a depth x is defined by

$$\left.\frac{\partial^k G(\epsilon,u;x)}{\partial u^k}\right|_{u=1}=\sum_{n_1=0}^\infty\sum_{n_2=0}^\infty n_1(n_1-1)\ldots(n_1-k+1)\Phi(\epsilon,n_1,n_2;x)=S^k(\epsilon,x).\qquad\ldots\ldots(32)$$

A formal solution for $S^k(\epsilon, x)$ follows immediately from the above since $\Phi(\epsilon, n_1, n_2; x)$ is given by (14), hence

$$S^k(\epsilon, x) = \sum_{n_1=0}^{\infty} \sum_{n_2=0}^{\infty} f_n(x)/n_2!\,(n_1-k)! \int_0^{\epsilon} \overset{n_2}{\cdots} \int_0^{\epsilon} \int_{\epsilon}^{1} \overset{n_1}{\cdots} \int_{\epsilon}^{1} L_n M_n(s_1, \ldots, s_n)\, d\epsilon_1 \ldots d\epsilon_n. \qquad \ldots\ldots(33)$$

It is obvious that to obtain explicit results for a given k, from (33), is by no means an easy task. In order to circumvent this difficulty we give below the diffusion equation obeyed by the kth moments and methods of solution which are more convenient for obtaining explicit results.

From (29), (30) and (32) we obtain

$$\frac{\partial}{\partial x} S^k(\epsilon, x) + S^k(\epsilon, x) = \int_0^{\infty} \int_0^{\infty} \sum_{l=0}^{k'} 2^{-\delta_{2l,k}} \binom{k}{l} S^l\left(\frac{\epsilon}{\epsilon_1}, x\right) S^{k-l}\left(\frac{\epsilon}{\epsilon_2}, x\right) \bar{w}(\epsilon_1, \epsilon_2)\, d\epsilon_1\, d\epsilon_2, \qquad \ldots\ldots(34)$$

where $k' = k/2$ when k is even and $(k-1)/2$ when k is odd. The exponent of the 2 gives the value $\frac{1}{2}$ when $2l = k$ and 1 at all other times. The solution of (34) for $k=1$ may be obtained by means of a single Mellin transform, thus:

$$S^1(\epsilon, x) = \frac{1}{2\pi i} \int_{s_0 - i\infty}^{s_0 + i\infty} \epsilon^{-s} \exp\{-\alpha(s) x\}\, ds/s. \qquad \ldots\ldots(35)$$

In a similar manner, using the solution (35) obtained for $k=1$, the solution for $k=2$ may be obtained. The procedure may then be repeated for $k=3$, etc. There is no need, however, to use this lengthy method, since the solution of (34) may be obtained directly. Assume that the solution is given by

$$S^k(\epsilon, x) = \frac{1}{(2\pi i)^k} \int_{s_1' - i\infty}^{s_1' + i\infty} \cdots \int_{s_k' - i\infty}^{s_k' + i\infty} \epsilon^{-(s_1 + \cdots + s_k)} Z_k(s_1, \ldots, s_k; x) \frac{ds_1}{s_1} \cdots \frac{ds_k}{s_k}. \qquad \ldots\ldots(36)$$

Substituting (36) into (34) yields the differential equation satisfied by the Z_k which can be shown to be identical with that satisfied by the n-fold Mellin transform of (27) when a given term is not repeated for all combinations of the k variables, i.e. for example, instead of

$$\{W(s_1, s_2) W(s_1 + s_2, s_3) + W(s_1, s_3) W(s_1 + s_3, s_2) + W(s_2, s_3) W(s_2 + s_3, s_1)\}$$

we write $3W(s_1, s_2) W(s_1 + s_2, s_3)$. The solution for $S^k(\epsilon, x)$ is hence given by

$$S^k(\epsilon, x) = \frac{1}{(2\pi i)^k} \int_{s_1' - i\infty}^{s_1' + i\infty} \cdots \int_{s_n' - i\infty}^{s_n' + i\infty} \epsilon^{-(s_1 + \cdots + s_k)} V_k(s_1, \ldots, s_k; x) \frac{ds_1}{s_1} \cdots \frac{ds_k}{s_k}. \qquad \ldots\ldots(37)$$

§6. DISCUSSION

The most striking feature of the solution of the fluctuation problem is this: any of the solutions for distribution functions or moments dealing with n particles invariably lead to an n-fold inverse transform. Once the form of the solution is known it is a relatively simple matter to find the exact solution. Furthermore, it is easier to solve for the most general distribution function itself, rather than for special cases of it. The methods of solution given in this paper are *directly* applicable to the electron–photon cascade. We shall, in the near future, present the solutions for the distribution functions in this case as well.

We have not attempted to discuss the numerical evaluation of any of the expressions given in our work. This we hope to do shortly. For small numbers of particles the evaluation appears straightforward.

ACKNOWLEDGMENT

I should like to express my thanks to Dr. J. W. Gardner for many helpful discussions during the early stage of this work.

APPENDIX

We give below the expressions for $M_2(s_1, s_2)$ and $M_4(s_1, s_2, s_3, s_4)$.

$$M_2(s_1, s_2) = 2W(s_1, s_2).$$

$$\begin{aligned}M_4(s_1, s_2, s_3, s_4) = \frac{2^3}{3!}\{&W(s_1, s_2)W(s_1+s_2, s_3)W(s_1+s_2+s_3, s_4) + W(s_1, s_2)W(s_1+s_2, s_4)W(s_1+s_2+s_4, s_3)\\ &+W(s_1, s_3)W(s_1+s_3, s_2)W(s_1+s_2+s_3, s_4) + W(s_1, s_3)W(s_1+s_3, s_4)W(s_1+s_3+s_4, s_2)\\ &+W(s_1, s_4)W(s_1+s_4, s_2)W(s_1+s_2+s_4, s_3) + W(s_1, s_4)W(s_1+s_4, s_3)W(s_1+s_3+s_4, s_2)\\ &+W(s_2, s_3)W(s_2+s_3, s_1)W(s_1+s_2+s_3, s_4) + W(s_2, s_3)W(s_2+s_3, s_4)W(s_2+s_3+s_4, s_1)\\ &+W(s_2, s_4)W(s_2+s_4, s_1)W(s_1+s_2+s_4, s_3) + W(s_2, s_4)W(s_2+s_4, s_3)W(s_2+s_3+s_4, s_1)\\ &+W(s_3, s_4)W(s_3+s_4, s_1)W(s_1+s_3+s_4, s_2) + W(s_3, s_4)W(s_3+s_4, s_2)W(s_2+s_3+s_4, s_1)\\ &+2W(s_1, s_2)W(s_3, s_4)W(s_1+s_2, s_3+s_4) + 2W(s_1, s_3)W(s_2, s_4)W(s_1+s_3, s_2+s_4)\\ &+2W(s_1, s_4)W(s_2, s_3)W(s_1+s_4, s_2+s_3)\}.\end{aligned}$$

The expressions for $V_2(s_1, s_2;\, x)$ and $V_3(s_1, s_2, s_3;\, x)$ may be obtained either from (23) or from (27) after an n-fold Mellin transform has been taken. It will be found that in practice it is easier to obtain explicit results from the latter case.

$$V_2(s_1, s_2;\, x) = \frac{2W(s_1, s_2)}{\alpha(s_1+s_2) - \alpha(s_1) - \alpha(s_2)}\{\exp[-\{\alpha(s_1) + \alpha(s_2)\}x] - \exp[-\alpha(s_1+s_2)x]\}$$

$$\begin{aligned}V_3(s_1, s_2, s_3;\, x) = {}& \frac{4W(s_1, s_2)W(s_1+s_2, s_3)}{\alpha(s_1) + \alpha(s_2) - \alpha(s_1+s_2)}\left\{\frac{\exp[-\{\alpha(s_1+s_2) + \alpha(s_3)\}x] - \exp[-\alpha(s_1+s_2+s_3)x]}{\alpha(s_1+s_2+s_3) - \alpha(s_1+s_2) - \alpha(s_3)}\right.\\ &\left. - \frac{\exp[-\{\alpha(s_1) + \alpha(s_2) + \alpha(s_3)\}x] - \exp[-\alpha(s_1+s_2+s_3)x]}{\alpha(s_1+s_2+s_3) - \alpha(s_1) - \alpha(s_2) - \alpha(s_3)}\right\}\\ &+ \frac{4W(s_1, s_3)W(s_1+s_3, s_2)}{\alpha(s_1) + \alpha(s_3) - \alpha(s_1+s_3)}\left\{\frac{\exp[-\{\alpha(s_1+s_3) + \alpha(s_2)\}x] - \exp[-\alpha(s_1+s_2+s_3)x]}{\alpha(s_1+s_2+s_3) - \alpha(s_1+s_3) - \alpha(s_2)}\right.\\ &\left. - \frac{\exp[-\{\alpha(s_1) + \alpha(s_2) + \alpha(s_3)\}x] - \exp[-\alpha(s_1+s_2+s_3)x]}{\alpha(s_1+s_2+s_3) - \alpha(s_1) - \alpha(s_2) - \alpha(s_3)}\right\}\\ &+ \frac{4W(s_2, s_3)W(s_2+s_3, s_1)}{\alpha(s_2) + \alpha(s_3) - \alpha(s_2+s_3)}\left\{\frac{\exp[-\{\alpha(s_2+s_3) + \alpha(s_1)\}x] - \exp[-\alpha(s_1+s_2+s_3)x]}{\alpha(s_1+s_2+s_3) - \alpha(s_2+s_3) - \alpha(s_1)}\right.\\ &\left. - \frac{\exp[-\{\alpha(s_1) + \alpha(s_2) + \alpha(s_3)\}x] - \exp[-\alpha(s_1+s_2+s_3)x]}{\alpha(s_1+s_2+s_3) - \alpha(s_1) - \alpha(s_2) - \alpha(s_3)}\right\}.\end{aligned}$$

REFERENCES

JÁNOSSY, L., 1949, *Proc. Phys. Soc.* A, **63,** 241.

JÁNOSSY, L., and MESSEL, H., 1951, *Proc. Roy. Irish Acad.*, A, **54,** 246.

MESSEL, H., 1951 a, *Proc. Roy. Irish Acad.* A, **54,** 125 ; 1951 b, *Commun. Dublin Inst. for Adv. Studies, Series* A, No. 7.

MESSEL, H., and POTTS, R. B., 1952, *Proc. Phys. Soc.* A, **65,** 473.

The Solution of the Fluctuation Problem in a Finite Absorber for Nucleon Cascades

By H. MESSEL and R. B. POTTS
Department of Mathematical Physics, University of Adelaide, South Australia

MS. received 12*th December* 1951

ABSTRACT. The complete analytical solution of the fluctuation problem for nucleon cascades developing in a finite absorber is given. Expressions are obtained for the general number distribution function and for the *n*th moments.

§ 1. INTRODUCTION

In a previous paper (Messel 1952, hereafter referred to as I) an analytical solution of the fluctuation problem for nucleon cascades developing in homogeneous nuclear matter was presented. The methods of solution developed in I were of general applicability and in the present work we apply them to solve the similar fluctuation problem in a finite absorber.

In §§ 2 and 3 of this paper we give the diffusion equations and their analytic solution for the number distribution function which expresses the probability of finding a stated number of particles with given energies at a given depth. From this distribution function one can in principle obtain all other number distribution functions of interest in the theory. An example is then discussed in detail for the case of four particles. In §§ 5–7 the diffusion equations for the *n*th moments and their solution are given. The actual evaluation of the expressions obtained is not attempted in the present paper but is left over to subsequent work, where the problem of evaluating the function appearing in the solution of the fluctuation problem both for nucleon and electron–photon cascade theory will be discussed.

Throughout our work we shall retain the same assumptions regarding the cross section for nucleon–nucleon collisions as in I.

PART I—THE DISTRIBUTION FUNCTION

§ 2. DIFFUSION EQUATIONS

Let $H_n(E_0; E_1, \ldots, E_n; \theta)\, dE_1 \ldots dE_n$ be the differential probability that after a depth θ (measured in units of the interaction mean free path) a primary nucleon of energy E_0 has given rise to n nucleons with energies in the ranges $E_j, E_j + dE_j, j = 1, \ldots n$ in any order; and let $Q_n(E_0; E_1, \ldots, E_n) dE_1 \ldots dE_n d\theta$ be the probability that a nucleon of energy E_0 collides with a nucleus in travelling a distance $d\theta$ and gives rise to n nucleons with energies in the ranges $E_j, E_j + dE_j$, $j = 1, \ldots, n$ in any order. Assuming nuclei to be spherical in shape

$$Q_n(E_0; E_1, \ldots, E_n) = \int_0^{D_A} F_n(E_0; E_1, \ldots, E_n; x) 2x\, dx / D_A^2, \quad \ldots\ldots(1)$$

where, as in I, $F_n(E_0; E_1, \ldots, E_n; x)\, dE_1 \ldots dE_n$ is defined as the differential probability that after a distance x in homogeneous nuclear matter a primary nucleon of energy E_0 has given rise to n nucleons with energies in the ranges $E_j, E_j + dE_j, j = 1, \ldots, n$ in any order. D_A is the diameter of a nucleus of atomic number A. The function F_n has been determined in I and shown to be a homogeneous function of the primary and secondary energies. Hence it follows that Q_n and therefore H_n are homogeneous functions as well.

From a consideration of all possible first nucleon–nucleus collisions follows the diffusion equation for H_n in the form

$$\frac{\partial}{\partial\theta}H_n + H_n = \sum_{v=1}^{n}\frac{1}{v!}\int_0^\infty dE_{10}\ldots\int_0^\infty dE_{v0}Q_v(E_0;E_{10},\ldots,E_{v0}) \times \sum_{n(v)}\prod_{j=1}^{v} H_{n(j)}(E_{j0};E_{j1},\ldots,E_{jn(j)};\theta), \quad \ldots\ldots(2)$$

where $\Sigma_{n(v)}$ signifies summation over all compositions of $E_1,\ldots,E_n$ into v groups $E_{11},\ldots,E_{1n(1)};\ldots;E_{v1},\ldots,E_{vn(v)}$ such that $\Sigma_{j=1}^{v}n(j)=n$ in all possible ways, $n(j)>0$. The order within each group of $n(j)$ terms is immaterial. There are just $n!/n(1)!\ldots n(v)!$ ways of choosing the v groups $n(j)$, $j=1,\ldots,v$ from the $E_1,\ldots,E_n$.

An alternative form of the diffusion equation may be obtained by considering all possible last collisions; in this case

$$\frac{\partial}{\partial\theta}H_n + nH_n = \sum_{k=1}^{n}\sum_{(k)}\int_0^\infty Q_k(\eta;E_1',\ldots,E_k')H_{n-k+1}(E_0;E_{k+1}',\ldots,E_n',\eta;\theta)d\eta, \quad \ldots\ldots(3)$$

where $\Sigma_{(k)}$ signifies summation over all compositions of the $E_1,\ldots,E_n$ into two groups $E_1',\ldots,E_k';E_{k+1}',\ldots,E_n'$. It is this form of the diffusion equation which will be used in finding the solution for H_n.

§ 3. SOLUTION OF THE DIFFUSION EQUATION

The partial integro-differential equations (3) can be transformed to a recurrence relation by taking an n-fold Mellin transform with respect to the energy variables and a single Laplace transform with respect to the depth dependence. Let

$$R_n(s_1,\ldots,s_n;p) = \int_0^\infty dE_1\ldots\int_0^\infty dE_n\int_0^\infty d\theta(E_1/E_0)^{s_1}\ldots(E_n/E_0)^{s_n}\,e^{-p\theta}H_n(E_0;E_1,\ldots,E_n;\theta) \quad \ldots\ldots(4)$$

and

$$N_n(s_1,\ldots,s_n) = \int_0^\infty dE_1\ldots\int_0^\infty dE_n(E_1/E_0)^{s_1}\ldots(E_n/E_0)^{s_n}Q_n(E_0;E_1,\ldots,E_n), \quad \ldots\ldots(5)$$

then eqn. (3) may be transformed into

$$(p+n)R_n = \sum_{k=1}^{n}\sum_{(k)}N_k(s_1',\ldots,s_k')R_{n-k+1}(s_{k+1}',\ldots,s_n',s_1'+\ldots+s_k';p),\ n>1. \quad \ldots\ldots(6)$$

From the initial condition $H_1(E_0;E;\theta=0)=\delta(E_0-E)$ and (3), (4) and (5) it may be shown that

$$R_1(s;p)=1/(p+h), \quad \ldots\ldots(7)$$

where

$$h=1-2\{1-(1+D_A)\exp(-D_A)\}/D_A^2. \quad \ldots\ldots(8)$$

The term on the right-hand side of (6) containing R_n for the case $k=1$ may be taken over to the left-hand side yielding

$$(p+nh)R_n = \sum_{k=2}^{n}\sum_{(k)}N_k(s_1',\ldots,s_k')R_{n-k+1}(s_{k+1}',\ldots,s_n',s_1'+\ldots+s_k';p),\ n>1. \quad \ldots\ldots(9)$$

The solution of the recurrence relation expressed by (9) may be given in the following compact form:

$$R_{n+1}=\frac{1}{p+h}\sum_{c}\prod_{j=m}^{1}\sum_{\text{comb}} N_{n(j)+1}(s_{q(j-1)+1},\ldots,s_{q(j)},s_{q(j)+1}+\ldots+s_{n+1})\{p+[q(j)+1]h\}^{-1}, \qquad \ldots\ldots(10)$$

where

(*a*) Σ_c signifies the sum over the 2^{n-1} compositions of n; c is the composition $n(1)$, $n(2)$, ..., $n(m)$. $\Sigma_{j=1}^{m} n(j)=n$, m is the number of nucleon–nucleus collisions, $n(j)+1$ gives the number of nucleons arising from the jth nucleon–nucleus collision.

(*b*) $q(j)=n(1)+\ldots+n(j)$, $q(0)=0$ and $q(m)=n$.

(*c*) $\sum_{\text{comb}}$ signifies summation over all combinations $C_{n(j)+1}^{q(j)+1}$ of the $q(j)+1$ symbols $s_1, s_2, \ldots, s_{q(j)}, s_{q(j)+1}+\ldots+s_{n+1}$ taken $n(j)+1$ at a time.

It will be found most expedient to carry out the summation Σ_c first, then the product $\Pi_{j=m}^{1}$ and finally to sum from the right.

The 2^{n-1} compositions of n describe the different ways in which a nucleon can, by collision with nuclei, give rise to the $n+1$ nucleons. If from the jth nucleon–nucleus collision $n(j)+1$ nucleons emerge and there are m collisions, then $\Sigma_{j=1}^{m} n(j)=n$; hence the composition $n(1),n(2), \ldots, n(m)$ describes a traverse in which the primary nucleon collides with a nucleus giving rise to $n(1)+1$ nucleons; one of these nucleons then collides with a nucleus giving rise to $n(2)+1$ nucleons, etc., until the mth collision in which $n(m)+1$ nucleons emerge. The total number of nucleons is then $n+1$.

It is evident from (10) that the p-dependent factor of R_{n+1} is

$$\Pi_{j=0}^{m} 1/\{p+[q(j)+1]h\}$$

and that this factor is the same for each composition c. The case R_4, i.e. $n=3$, will be discussed in detail in the following section.

The solution for H_n is now immediate:

$$H_n(E_0; E_1, \ldots, E_n; \theta)=\mathbf{I}_n R_n(s_1, \ldots, s_n; p), \qquad \ldots\ldots(11)$$

where $\mathbf{I}_n$ is the inverse Laplace–Mellin transform operator:

$$\mathbf{I}_n=\frac{1}{(2\pi i)^{n+1}}\,E_0^{-n}\int_{u_1-i\infty}^{u_1+i\infty} ds_1 \ldots\ldots \int_{u_n-i\infty}^{u_n+i\infty} ds_n \int_{\lambda-i\infty}^{\lambda+i\infty} dp \left(\frac{E_0}{E_1}\right)^{s_1+1} \ldots \left(\frac{E_0}{E_n}\right)^{s_n+1} e^{p\theta}. \qquad \ldots\ldots(12)$$

From the distribution function H_n whose solution is given by (11) all other number distribution functions of interest in the theory may easily be obtained. For instance the probability of finding n_1 particles with energies greater than E and n_2 particles with energies less than E at a depth θ due to a primary of energy E_0 may be obtained by integrating over n_1 of the variables of H_n from E to ∞ and the remaining n_2 from 0 to E; hence

$$\phi(E_0; E, n_1, n_2; \theta)=\frac{1}{n_1!\,n_2!}\int_E^\infty \overset{n_1}{\cdots} \int_E^\infty \int_0^E \overset{n_2}{\cdots} \int_0^E H_n\, dE_1 \ldots dE_n. \qquad \ldots\ldots(13)$$

The physically interesting quantity is, however, the probability of finding n_1 particles with energies greater than E and an arbitrary number of particles with energies less than E at a depth θ, which is given by

$$\Phi(E_0, E, n_1; \theta) = \sum_{n_2=0}^{\infty} \frac{1}{n_1!\, n_2!} \int_E^{\infty} \overset{n_1}{\cdots} \int_E^{\infty} \int_0^E \overset{n_2}{\cdots} \int_0^E H_n \, dE_1 \ldots dE_n. \qquad \ldots\ldots(14)$$

The integrations in (13) and (14) over the energy variables are easily performed since the function H_n is a homogeneous function of the primary and secondary energies as seen from the solution given in (11) and (12).

§ 4. THE CASE R_4 $(s_1, s_2, s_3, s_4; p)$

It is evident that the solution for H_n given by (10), (11) and (12) is by no means elementary. In view of this we have thought it advisable to discuss one case in full, the case R_4.

First it is necessary to list the compositions of $n=3$; there are $2^2=4$ in all:

Composition	m	$n(j)+1$	Form
$c(1)=1,1,1$	3	2,2,2	as fig. 1 (*a*), (*b*)
$c(2)=1,2$	2	2,3	as fig. 2.
$c(3)=2,1$	2	3,2	as fig. 3.
$c(4)=3$	1	4	as fig. 4.

The values of m in the second column give the number of nucleon–nucleus collisions, the values of $n(j)+1$ give the numbers of nucleons emerging from the m nucleon–nucleus collisions, and the diagrams represent possible ways in which the traverse can occur. These will be discussed further below.

For the composition $c(1)$ the corresponding term in expression (10) is

$$\begin{aligned}
&\frac{1}{(p+h)(p+2h)(p+3h)(p+4h)} \sum_{C_2^4} N_2(s_3, s_4) \sum_{C_2^3} N_2(s_2+s_3+s_4) \\
&\qquad\qquad \sum_{C_2^2} N_2(s_1, s_2+s_3+s_4) \\
&= \frac{1}{(p+h)(p+2h)(p+3h)(p+4h)} \sum_{C_2^4} N_2(s_3, s_4)\{N_2(s_2, s_3+s_4)N_2(s_1, s_2+s_3+s_4) \\
&\qquad\qquad + N_2(s_1, s_3+s_4)N_2(s_2, s_1+s_3+s_4) \\
&\qquad\qquad + N_2(s_2, s_1)N_2(s_3+s_4, s_2+s_1)\}, \qquad \ldots\ldots(15)
\end{aligned}$$

where $\Sigma_{C_2^4}$ signifies summation over all combinations of s_1, s_2, s_3, s_4 taken two at a time. When this last summation is carried out the result becomes entirely analogous to the expression for $M_4(s_1, s_2, s_3, s_4)$ given for homogeneous nuclear matter in the appendix of I. This is to be expected since the composition $c(1)=1, 1, 1$ corresponds to a traverse in which each nucleon–nucleus collision gives rise to two nucleons just as in nucleon–nucleon collisions.

It is interesting to notice that the terms in (15) after the last summation has been performed can be represented by 'trees' of the type given in fig. 1. The terms are constructed from the trees by reading from bottom to top and adding the s variables at each collision. The two trees given in fig. 1 correspond to the terms

$$N_2(s_3, s_4)N_2(s_2, s_3+s_4)N_2(s_1, s_2+s_3+s_4) \text{ and } N_2(s_3, s_4)N_2(s_1, s_2)N_2(s_3+s_4, s_1+s_2).$$

The remaining terms in (15) may be obtained from fig. 1 by means of a simple permutation of the variables.

The p-dependent factor in (15) may also be read off from the tree, fig. 1 (*a*). Reading from top to bottom, there are at first one, two, then three and finally four branches at each level; the corresponding p-dependent factor is $1/(p+h)(p+2h)(p+3h)(p+4h)$. These factors are always determined by trees of the type in fig. 1 (*a*) (and not 1 (*b*)). It is known that the factors depend only on the composition, and there is a tree of this type for every composition.

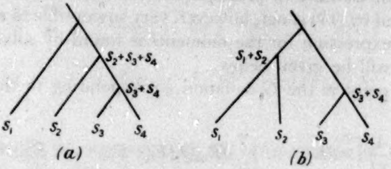


Fig. 1.

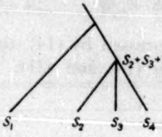


Fig. 2.

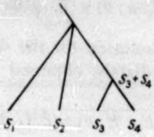


Fig. 3.

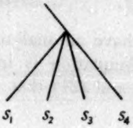


Fig. 4.

For the composition $c(2)$ the corresponding term in (10) is

$$\begin{aligned}&\frac{1}{(p+h)(p+2h)(p+4h)}\sum_{C_3^4} N_3(s_2, s_3, s_4)\sum_{C_2^2} N_2(s_1, s_2+s_3+s_4)\\ &=\frac{1}{(p+h)(p+2h)(p+4h)}\{N_3(s_2, s_3, s_4)N_2(s_1, s_2+s_3+s_4)\\ &\quad+N_3(s_3, s_4, s_1)N_2(s_2, s_3+s_4+s_1)+N_3(s_4, s_1, s_2)N_2(s_3, s_4+s_1+s_2)\\ &\quad+N_3(s_1, s_2, s_3)N_2(s_4, s_1+s_2+s_3)\}. \qquad \ldots\ldots(16)\end{aligned}$$

The tree from which these terms may be obtained is given in fig. 2.

For the composition $c(3)$ the term is

$$\begin{aligned}&\frac{1}{(p+h)(p+3h)(p+4h)}\sum_{C_2^4} N_2(s_3, s_4)\sum_{C_3^3} N_3(s_1, s_2, s_3+s_4)\\ &=\frac{1}{(p+h)(p+3h)(p+4h)}\{N_2(s_3, s_4)N_3(s_1, s_2, s_3+s_4)\\ &\quad+N_2(s_4, s_1)N_3(s_2, s_3, s_4+s_1)+N_2(s_1, s_2)N_3(s_3, s_4, s_1+s_2)\\ &\quad+N_2(s_2, s_3)N_3(s_4, s_1, s_2+s_3)+N_2(s_1, s_3)N_3(s_2, s_4, s_1+s_3)\\ &\quad+N_2(s_2, s_4)N_3(s_1, s_3, s_2+s_4)\}, \qquad \ldots\ldots(17)\end{aligned}$$

corresponding to the tree given in fig. 3.

Finally the composition $c(4)$ yields the term

$$\{1/(p+h)(p+4h)\}N_4(s_1, s_2, s_3, s_4), \qquad \ldots\ldots(18)$$

and the tree representing it is given in fig. 4.

The solution for R_4 is now obtained by adding the terms given by (15), (16), (17) and (18).

PART II—THE MOMENTS

§ 5. DIFFUSION EQUATION FOR THE MOMENTS

The nth factorial moment of the distribution function Φ found in (14) is defined to be

$$T_n(E_0; E; \theta) = \sum_{k=0}^{\infty} \frac{k!}{(k-n)!} \Phi(E_0; E, k; \theta). \qquad \ldots\ldots(19)$$

Since Φ is known, we have in principle the solution for the T_n. The solution for T_n as expressed by (19) is not, however, very susceptible to explicit evaluation. A more suitable expression for the moments is found by solving their diffusion equation, which will be given below.

The starting point is the G-equation corresponding to the function Φ (see Messel 1951):

$$\frac{\partial}{\partial\theta} G + G = \sum_{r=1}^{\infty} \frac{1}{r!} \int_0^{\infty} dE_{10} \ldots \int_0^{\infty} dE_{r0} Q_r(E_0; E_{10}, \ldots, E_{r0}) \prod_{j=1}^{r} G(E_{j0}; E, u; \theta), \qquad \ldots\ldots(20)$$

where

$$G(E_0; E; u; \theta) = \sum_{k=0}^{\infty} u^k \Phi(E_0; E, k; \theta). \qquad \ldots\ldots(21)$$

Since we have an analytical solution for the Φ expressed by (14), the solution for the G-equation is immediately obtained from (14) and (21). It follows from (19) and (21) that

$$T_n(E_0; E; \theta) = (\partial^n G/\partial u^n)|_{u=1}. \qquad \ldots\ldots(22)$$

By successive differentiation of (20) and making use of the relation (22) and the fact that $G(E_0; E, u=1; \theta) = 1$, we find the diffusion equation satisfied by T_n:

$$\frac{\partial}{\partial\theta} T_n + T_n = \sum_{r=1}^{\infty} \frac{1}{r!} \int_0^{\infty} dE_{10} \ldots \int_0^{\infty} dE_{r0} Q_r(E_0; E_{10}, \ldots, E_{r0})$$
$$\times \Sigma^{(r)} \frac{n!}{n(1)! \ldots n(r)!} \prod_{j=1}^{r} T_{n(j)}(E_{j0}; E; \theta), \qquad \ldots\ldots(23)$$

where $\Sigma^{(r)}$ signifies summation over all compositions of n into r groups $n(j)$, $j = 1, \ldots, r$ ($n(j)$ may be zero).

The solution of eqn. (23) for the cases $n = 1$ and 2 has previously been obtained (Messel 1951). One could obtain the higher moments by successive substitutions into eqn. (23). This proves to be a very lengthy task for values of $n > 3$. Equation (23) may however be solved in a manner analogous to that given in I for homogeneous nuclear matter. In order to do so, we must first consider the solution of a subsidiary diffusion equation, given below.

§ 6. DIFFUSION EQUATION FOR J_n

Let $J_n(E_0; E_1, \ldots, E_n; \theta)\, dE_1 \ldots . dE_n$ be the probability of finding n particles with energies in the range E_j, $E_j + dE_j$ at a depth θ, and any number of particles with arbitrary energies, due to a primary of energy E_0. As for the case of the H_n, two separate diffusion equations may be set up.

By considering all possible first collisions we find

$$\frac{\partial}{\partial\theta} J_n + J_n = \sum_{r=1}^{\infty} \frac{1}{r!} \int_0^{\infty} dE_{10} \ldots \int_0^{\infty} dE_{r0} Q_r(E_0; E_{10}, \ldots, E_{r0})$$
$$\times \sum_{n(r)} \prod_{j=1}^{r} J_{n(j)}(E_{j0}; E_{j1}, \ldots, E_{jn(j)}; \theta), \qquad \ldots\ldots(24)$$

where $\Sigma_{n(r)}$ signifies summation over all compositions of $E_1, \ldots, E_n$ into r groups $E_{11}, \ldots, E_{1n(1)}; \ldots; E_{r1}, \ldots, E_{rn(r)}$, such that $\Sigma_{j=1} n(j) = n$ and where $n(j)$ may be zero. There are just $n!/n(1)! \ldots n(r)!$ ways of dividing the $E_1, \ldots, E_n$ into the r groups.

An alternative form of the diffusion equation is obtained by considering all possible last collisions; in this case we find

$$\frac{\partial}{\partial\theta} J_n + nJ_n = \sum_{k=1}^{n} \sum_{r=k}^{\infty} \sum_{(k)} \frac{1}{(r-k)!} \int_0^\infty d\eta \int_0^\infty d\xi_1 \ldots \int_0^\infty d\xi_{r-k}$$
$$\times Q_r(\eta; E_1', \ldots, E_k', \xi_1, \ldots, \xi_{r-k}) J_{n-k+1}(E_0; E'_{k+1}, \ldots, E_n', \eta; \theta), \qquad \ldots\ldots(25)$$

where $\Sigma_{(k)}$ signifies the summation over all compositions of $E_1, \ldots, E_n$ into two groups $E_1', \ldots, E'_{n-k}$ and $E'_{n-k+1}, \ldots, E_n'$.

Let us now define

$$D_n(E_0; E_1, \ldots, E_n) = \int_0^{D_A} C_n(E_0; E_1, \ldots, E_n; x) 2x\,dx/D_A^2, \qquad \ldots\ldots(26)$$

where from I

$$C_n(E_0; E_1, \ldots, E_n; x) = \sum_{r=0}^{\infty} \frac{1}{r!} \int_0^\infty dE_{n+1} \ldots \int_0^\infty dE_{n+r} F_{n+r}(E_0; E_1, \ldots, E_{n+r}; x); \qquad \ldots\ldots(27)$$

hence using (1)

$$D_n(E_0; E_1, \ldots, E_n) = \sum_{r=0}^{\infty} \frac{1}{r!} \int_0^\infty dE_{n+1} \ldots \int_0^\infty dE_{n+r} Q_{n+r}(E_0; E_1, \ldots, E_{n+r}). \qquad \ldots\ldots(28)$$

The function D_n expresses the probability that a primary nucleon of energy E_0 on colliding with a nucleus will give rise to n nucleons with energies in the intervals dE_j, $j = 1, \ldots, n$, and any number of nucleons with arbitrary energies. In I the solution for C_n was left in the form of a recurrence relation. It may be shown that the explicit solution for C_n is

$$C_n = \mathbf{I}_n \overline{V}_n, \qquad \ldots\ldots(29)$$

where $\overline{V}_{n+1}(s_1, \ldots, s_{n+1}; p) = \dfrac{1}{p + \alpha(s_1 + \ldots + s_{n+1})} \prod_{j=n}^{1} \sum_{\text{comb}}$

$$W(s_{\alpha(j)}, s_{\alpha(j)+1} + \ldots + s_{n+1}) \{p + \alpha(s_1) + \ldots + \alpha(s_{\alpha(j)}) + \alpha(s_{\alpha(j)+1} + \ldots + s_{n+1})\}^{-1} \qquad \ldots\ldots(30)$$

and the notation is the same as in (10) with the composition $n(1) = n(2) = \ldots = n(n) = 1$. We refer the readers to I for the definitions of α and W.

Now since C_n is known, the value of D_n may be obtained by performing the x integration as in (26).

Using the relation (28) for D_n, eqn. (25) may be reduced to the form

$$\frac{\partial}{\partial\theta} J_n + nJ_n = \sum_{k=1}^{n} \sum_{(k)} \int_0^\infty D_k(\eta; E_1', \ldots, E_k') J_{n-k+1}(E_0; E'_{k+1}, \ldots, E_n', \eta; \theta)\, d\eta. \qquad \ldots\ldots(31)$$

The solution for J_n may now be obtained from (31) by applying a single Laplace and an n-fold Mellin transform and solving the resultant equation in a manner similar to that used to find H_n. We find

$$J_n = \mathbf{I}_n Y_n. \qquad \ldots\ldots(32)$$

$$Y_{n+1}(s_1, \ldots, s_{n+1}; p) = \frac{1}{p + h(s_1 + \ldots + s_{n+1})} \sum_{c} \prod_{j=m}^{1} \sum_{\text{comb}} B_{n(j)+1}$$
$$\times (s_{\sigma(j-1)+1}, \ldots, s_{\sigma(j)}, s_{\sigma(j)+1} + \ldots + s_{n+1})$$
$$\times \{p + h(s_1) + \ldots + h(s_{\sigma(j)}) + h(s_{\sigma(j)+1} + \ldots + s_{n+1})\}^{-1}, \qquad \ldots\ldots(33)$$

where B_n is the n-fold Mellin transform of D_n and

$$h(s) = 1 - 2[1 - \{1 + D_A \alpha(s)\} \exp\{-D_A \alpha(s)\}] / \{D_A \alpha(s)\}^2. \qquad \ldots\ldots(34)$$

The terms which occur in (33) can be constructed from trees just as for (10). The function B_n occurs in exactly the same way as the function N_n but the p-dependent factors are now different for each term in Σ_{comb}. They can, however, be read off from the tree. For example, fig. 2 corresponds to the term

$$B_3(s_2, s_3, s_4) B_2(s_1, s_2 + s_3 + s_4) \{[p + h(s_1 + s_2 + s_3 + s_4)]$$
$$\times [p + h(s_1) + h(s_2 + s_3 + s_4)][p + h(s_1) + h(s_2) + h(s_3) + h(s_4)]\}^{-1}$$

in the expression for Y_4. The p-dependent factors are written down from the tree by reading from top to bottom and at each level choosing as the variables of h the values of s on the branches.

§7. THE SOLUTION FOR T_n

We are now in position to give the solution for T_n. Assume the solution to be

$$T_n(E_0; E; \theta) = \frac{1}{(2\pi i)^n} \int_{u_1 - i\infty}^{u_1 + i\infty} \ldots \int_{u_n - i\infty}^{u_n + i\infty} \left(\frac{E}{E_0}\right)^{-(s_1 + \ldots + s_n)}$$
$$\times Z_n(s_1, \ldots, s_n; \theta) \frac{ds_1}{s_1} \ldots \frac{ds_n}{s_n}. \qquad \ldots\ldots(35)$$

It remains for us to determine $Z_n(s_1, \ldots, s_n; \theta)$. Substitute the value of T_n given in (35) into eqn. (23). The resultant differential equation for Z_n can then be shown to be *identical* with that satisfied by the n-fold Mellin transform of eqn. (24) for J_n, when a given term is not repeated for all combinations of the n-variables s_j, $j = 1, \ldots, n$. Hence the solution for T_n is given by

$$T_n(E_0; E; \theta) = \frac{1}{(2\pi i)^n} \int_{u_1 - i\infty}^{u_1 + i\infty} \ldots \int_{u_n - i\infty}^{u_n + i\infty} \left(\frac{E}{E_0}\right)^{-(s_1 + \ldots + s_n)}$$
$$\times \bar{Y}_n(s_1, \ldots, s_n; \theta) \frac{ds_1}{s_1} \ldots \frac{ds_n}{s_n}, \qquad \ldots\ldots(36)$$

where $$\bar{Y}_n(s_1, \ldots, s_n; \theta) = \frac{1}{2\pi i} \int_{\lambda - i\infty}^{\lambda + i\infty} e^{p\theta} Y_n(s_1, \ldots, s_n; p)\, dp. \qquad \ldots\ldots(37)$$

This completes the solution of the fluctuation problem in nucleon cascade theory. There only remains the explicit evaluation of the different analytical expressions arising above. We hope to consider this problem in some detail in the near future.

REFERENCES

MESSEL, H., 1951, *Commun. Dublin Inst. for Advanced Studies, Series* A, No. 7 ; 1952, *Proc. Phys. Soc.* A, **65**, 465.

On the Scattering of Fast Electrons by Nuclei

By L. R. B. ELTON
Wheatstone Department of Physics, King's College, London

Communicated by H. S. W. Massey; MS. received 7th February 1952

ABSTRACT. It is shown that, contrary to expectations from the non-relativistic case, the phase-shift ζ_n between the partial waves of electrons scattered by a point nucleus and by an extended nucleus respectively is positive for sufficiently large n, and that this is a spin effect and not merely a relativistic one. An approximate formula for ζ_n is evolved and its usefulness is discussed.

---

§ 1. INTRODUCTION

Recent calculations, by Elton (1950, to be referred to as I) and Acheson (1951) on the one hand and by Parzen (1950 b) on the other, of the scattering of high energy electrons by a point and extended nucleus respectively have shown up an apparent disagreement, in that Parzen finds that the phase shift ζ_n (his δ_l) between the partial waves for the two cases tends to π as n tends to ∞, while Acheson and the author assumed that it tends to zero monotonically. Since we both find that $\zeta_0<0$, Parzen concludes that $\tan\zeta_n<0$ for small n, but is greater than 0 and tending to $+0$ for large n, while Acheson and the author assumed that $\tan\zeta_n<0$ for all n and tends to -0 monotonically for large n. It will be shown now that neither view is correct, in that $\zeta_n\to 0$ and $\tan\zeta_n\to +0$ as $n\to\infty$. It follows from this result that phase shifts ζ_n have to be calculated up to values of n for which $\zeta_n>0$ and negligible, since a negligibly small negative value of ζ_n may be followed by a larger positive one. The calculations in I were therefore taken a stage further and ζ_2 was calculated.

The notation of I will be followed throughout in this paper.

§ 2. PROOF THAT $\zeta_n\to 0$

As Parzen has pointed out (1950 b, eqn. (17)), $\theta_n\to 0$ as $n\to\infty$, where θ_n is the phase difference between the partial waves corresponding to the regular and irregular solutions for the point nucleus, and hence it follows from I(12) that $\tan\zeta_n\to 0$. To show that $\zeta_n\to 0$ we prove that for large n the functions $\mathscr{G}_n(r)$ and $\mathscr{K}_n(r)$ which are the regular solutions of the radial equation for a point nucleus and an extended nucleus respectively, have no zeros inside the nuclear radius, and that therefore on the nuclear radius they cannot be out of phase by as much as π.

Both $\mathscr{G}_n$ and $\mathscr{K}_n$ satisfy the differential equation I (8)

$$\mathscr{S}''=f_n(r)\mathscr{S}, \qquad \ldots\ldots(1)$$

where

$$f_n(r)=-k^2+\frac{n(n+1)}{r^2}+\frac{2WV}{\hbar^2\mathbf{c}^2}-\frac{V^2}{\hbar^2\mathbf{c}^2}-\frac{n+1}{r}\frac{\alpha'}{\alpha}+\frac{3}{4}\frac{\alpha'^2}{\alpha^2}-\frac{1}{2}\frac{\alpha''}{\alpha},$$

and $\alpha=(W-V+\boldsymbol{m}\mathbf{c}^2)/\hbar\mathbf{c}$. Therefore, provided $f_n(r)>0$, $\mathscr{S}''>0$ as long as $\mathscr{S}>0$, i.e. $\mathscr{S}$ increases as long as $\mathscr{S}>0$; and since from the power series expansion of $\mathscr{S}$ it is clear that for sufficiently small r, $\mathscr{S}$ and $\mathscr{S}'$ have the same sign and can therefore be taken positive, it follows that $\mathscr{S}$ is positive and increasing as long as $f_n(r)>0$. Now for large n and $r\neq 0$, but small, the dominant term of $f_n(r)$ is clearly $n(n+1)/r^2$, and as r increases, all other terms of $f_n(r)$ remain small compared with $n(n+1)/r^2$ except possibly the first and third. Hence $f_n(r)$ vanishes first when r is approximately given by $(1-2WV/\hbar^2\mathbf{c}^2k^2)k^2r^2=n(n+1)$, i.e. when $kr=\mathrm{O}(n)$. Hence $\mathscr{G}_n$ and $\mathscr{K}_n$ do not vanish until $kr=\mathrm{O}(n)$, which proves the proposition.

§ 3. PROOF THAT $\tan \zeta_n \to +0$

It is next shown that for sufficiently large n, $\tan \zeta_n>0$. In this proof affixes P and E denote quantities referring to a point nucleus and to an extended nucleus respectively.

Equation (1) can be re-written for an extended nucleus

$$\frac{d^2\mathscr{S}}{dr^2}+\left[k^2-\frac{n(n+1)}{r^2}\,U_n{}^{\mathrm{P}}(r)\right]\mathscr{S}=V_n(r)\mathscr{S}, \qquad \ldots\ldots(2)$$

where $U_n(r)=f_n(r)+k^2-n(n+1)/r^2$ and $V_n(r)=U_n{}^{\mathrm{E}}(r)-U_n{}^{\mathrm{P}}(r)$. Thus $V_n(r)=0$ for $r>R$, the nuclear radius. We choose a solution $\mathscr{K}_n$ which satisfies the following conditions:

$$\mathscr{K}_n(0)=0, \quad \mathscr{K}_n\sim\text{const.}\exp\{i(kr+\gamma\log 2kr)\}, \quad \gamma=Z\mathbf{e}^2/\hbar v. \qquad \ldots\ldots(3)$$

Let $\mathscr{G}_n$ and $\mathscr{I}_n$ be the solutions of the equation

$$\frac{d^2\mathscr{S}}{dr^2}+\left[k^2-\frac{n(n+1)}{r^2}-U_n{}^{\mathrm{P}}(r)\right]\mathscr{S}=0, \qquad \ldots\ldots(4)$$

which satisfy the following conditions:

$$\begin{gathered}\mathscr{G}_n(0)=0, \quad \mathscr{G}_n\sim\mu^{-1/2}\sin\left(kr+\gamma\log 2kr-\tfrac{1}{2}n\pi+\eta_n{}^{\mathrm{P}}\right),\\ \mathscr{I}_n\sim\mu^{1/2}k^{-1}\exp\{i(kr+\gamma\log 2kr-\tfrac{1}{2}n\pi+\eta_n{}^{\mathrm{P}})\},\end{gathered} \qquad \ldots\ldots(5)$$

where $\mu=(W+\mathbf{mc}^2)/\hbar\mathbf{c}$. Then following Mott and Massey (1949) we have for all r

$$\mathscr{I}_n\frac{d\mathscr{G}_n}{dr}-\mathscr{G}_n\frac{d\mathscr{I}_n}{dr}=1 \qquad \ldots\ldots(6)$$

and hence $$\mathscr{K}_n=\mathscr{G}_n\int_\infty^r \mathscr{I}_n V_n \mathscr{K}_n\,dr-\mathscr{I}_n\int_0^r \mathscr{G}_n V_n \mathscr{K}_n\,dr. \qquad \ldots\ldots(7)$$

To this solution we must add $\mathscr{G}_n$, since when $V_n(r)=0$, $\mathscr{K}_n$ must reduce to $\mathscr{G}_n$. Therefore

$$\begin{aligned}\mathscr{K}_n\sim{}&\mu^{-1/2}\sin\left(kr+\gamma\log 2kr-\tfrac{1}{2}n\pi+\eta_n{}^{\mathrm{P}}\right)\\ &-\mu^{1/2}k^{-1}\exp\{i(kr+\gamma\log 2kr-\tfrac{1}{2}n\pi+\eta_n{}^{\mathrm{P}})\}\int_0^\infty \mathscr{G}_n V_n \mathscr{K}_n\,dr.\end{aligned} \qquad \ldots\ldots(8)$$

Now

$$\begin{aligned}\mathscr{K}_n&\sim\mu^{-1/2}\sin\left(kr+\gamma\log 2kr-\tfrac{1}{2}n\pi+\eta_n{}^{\mathrm{E}}\right) & \ldots\ldots(9)\\ &=\mu^{-1/2}\exp(-i\zeta_n)[\sin\left(kr+\gamma\log 2kr-\tfrac{1}{2}n\pi+\eta_n{}^{\mathrm{P}}\right)\\ &\quad+\{\exp(2i\zeta_n)-1\}\exp\{i(kr+\gamma\log 2kr-\tfrac{1}{2}n\pi+\eta_n{}^{\mathrm{P}})\}], & \ldots\ldots(10)\end{aligned}$$

where $\zeta_n=\eta_n{}^{\mathrm{E}}-\eta_n{}^{\mathrm{P}}$. Comparing (8) and (10) we have, after a little rearrangement,

$$\sin\zeta_n=-\frac{\mu}{k}\int_0^\infty \mathscr{G}_n V_n \mathscr{K}_n\,dr. \qquad \ldots\ldots(11)$$

Now for large n, ζ_n is small and hence to first order $\mathscr{K}_n$ can be replaced by $\mathscr{G}_n$ in the integral. Also $V_n(r)=0$ for $r>R$. We thus obtain the approximate formula

$$\zeta_n \simeq -\frac{\mu}{k}\int_0^R \mathscr{G}_n V_n \mathscr{G}_n \, dr. \qquad \ldots\ldots(12)$$

Therefore, for large n, the sign of ζ_n is opposite to that of $V_n(r)$. But

$$V_n(r)=\frac{1}{\hbar^2\mathbf{c}^2}(V_{\mathrm{E}}-V_{\mathrm{P}})(2W-V_{\mathrm{E}}-V_{\mathrm{P}})+\frac{n+1}{r}\left(\frac{\alpha_{\mathrm{P}}'}{\alpha_{\mathrm{P}}}-\frac{\alpha_{\mathrm{E}}'}{\alpha_{\mathrm{E}}}\right)$$
$$+\frac{3}{4}\left(\frac{\alpha_{\mathrm{E}}'^2}{\alpha_{\mathrm{E}}^2}-\frac{\alpha_{\mathrm{P}}'^2}{\alpha_{\mathrm{P}}^2}\right)-\frac{1}{2}\left(\frac{\alpha_{\mathrm{E}}''}{\alpha_{\mathrm{E}}}-\frac{\alpha_{\mathrm{P}}''}{\alpha_{\mathrm{P}}}\right),$$

and since all terms are $O(r^{-2})$ near the origin, it is clear that for an attractive potential ζ_n will have the same sign as $\alpha_{\mathrm{P}}\alpha_{\mathrm{E}}'-\alpha_{\mathrm{E}}\alpha_{\mathrm{P}}'$ for sufficiently large n. Now $\alpha_{\mathrm{P}}=\mu+\lambda/r$, where $\lambda=\gamma v/\mathbf{c}$, and we can write $\alpha_{\mathrm{E}}=\mu+\lambda g(r)$, where for any physically likely charge distribution $g(r)$ decreases and $rg(r)$ increases monotonically with r, and $0<g(r)<r^{-1}$, all for $r<R$. Then for $r<R$,

$$g'<0, \quad rg'+g>0; \quad \text{i.e.} \quad |g'|<g/r<1/r^2.$$

Thus the force at any point is less than the force due to the corresponding point nucleus. (Note that the converse of this, i.e. that if $|g'|<r^{-2}$, then $rg(r)$ increases monotonically with r, is not true.) Therefore

$$\begin{aligned}\alpha_{\mathrm{P}}\alpha_{\mathrm{E}}'-\alpha_{\mathrm{E}}\alpha_{\mathrm{P}}' &=(\mu+\lambda/r)\lambda g'+(\mu+\lambda g)\lambda/r^2\\ &=\lambda[\mu(g'+1/r^2)+\lambda r^{-1}(g'+g/r)]\\ &>0,\end{aligned}$$

using the above inequalities. Thus $\zeta_n>0$ for sufficiently large n.

This rather surprising result is peculiar to the Dirac equation, since for the Klein–Gordon equation, $V_n(r)=(V_{\mathrm{E}}-V_{\mathrm{P}})(2W-V_{\mathrm{E}}-V_{\mathrm{P}})/\hbar^2\mathbf{c}^2$, and for the Schrödinger equation, $V_n(r)=2\mathbf{m}(V_{\mathrm{E}}-V_{\mathrm{P}})/\hbar^2$, and in both these cases $\zeta_n<0$ for all n since $V_{\mathrm{E}}\geqslant V_{\mathrm{P}}$ for all r. Thus the effect is not a merely relativistic one, but one due to the spin of the electron.

§ 4. USE OF THE APPROXIMATE FORMULA

The accuracy of (12) has been checked by comparing values of ζ_n obtained from it with the exact values, for electrons of energy $40\mathbf{mc}^2$ being scattered by gold nuclei. The values are given in the table and it is clear that even for ζ_0 the approximate formula gives good results under these conditions (B and C refer to uniform and shell nucleus respectively). Lastly ζ_2^{C} was calculated and it was found that $\zeta_2^{\mathrm{C}}=-0{\cdot}00002$. Thus ζ_2^{C} is still negative, but it is clear from the trend of ζ_n that all further phase shifts can be neglected.

Phase shifts ζ_n^{B}, ζ_n^{C} obtained by exact calculations and from the approximate formula (12)

Phase shift	ζ_0^{B}	ζ_0^{C}	ζ_1^{B}	ζ_1^{C}
Exact	−0·196	−0·271	−0·002	−0·004
Approximate	−0·231	−0·321	−0·002	−0·004

The approximate formula will be particularly useful in calculating the effect of different models, once the phase for the spherical shell model has been calculated exactly. This is easiest to calculate since $\mathscr{K}_n^{\mathrm{C}}$ is a Bessel function.

Further, since in that case $\mathscr{K}_n{}^{\mathrm{C}}$ is known, the phase shifts for other models can be obtained from the formulae

$$\zeta_n{}^{\mathrm{E}}-\zeta_n{}^{\mathrm{C}}\simeq-\frac{\mu}{k}\int_0^R\mathscr{G}_n(U_n{}^{\mathrm{E}}-U_n{}^{\mathrm{C}})\mathscr{G}_n\,dr \qquad\ldots\ldots(13)$$

and

$$\zeta_n{}^{\mathrm{E}}-\zeta_n{}^{\mathrm{C}}\simeq-\frac{\mu}{k}\int_0^R\mathscr{K}_n{}^{\mathrm{C}}(U_n{}^{\mathrm{E}}-U_n{}^{\mathrm{C}})\mathscr{K}_n{}^{\mathrm{C}}\,dr, \qquad\ldots\ldots(14)$$

where the affix E refers to any model for an extended nucleus, other than the shell model, and the correct result must clearly lie between the results obtained from (13) and (14). This method is likely to give more accurate results than the variation principle evolved by Parzen (1950 a) for the same purpose, since Parzen in effect replaces $\mathscr{G}_n(kr)$ or $\mathscr{K}_n{}^{\mathrm{C}}(kr)$ by the less accurate Bessel function $J_n(kr)$ and neglects squares of the potential. A numerical check has been carried out, by calculating $\zeta_0{}^{\mathrm{E}}-\zeta_0{}^{\mathrm{C}}$ by Parzen's method and from formula (13), which confirms the superior accuracy of the present method.

ACKNOWLEDGMENT

My thanks are due to Mr. L. Castillejo for many helpful discussions.

REFERENCES

Acheson, L. K., 1951, *Phys. Rev.*, **82**, 488.
Elton, L. R. B., 1950, *Proc. Phys. Soc.* A, **63**, 1115.
Mott, N. F., and Massey, H. S. W., 1949, *The Theory of Atomic Collisions* (Oxford: University Press, 2nd edition), chap. VI.
Parzen, G., 1950 a, *Phys. Rev.*, **80**, 261; 1950 b, *Ibid.*, **80**, 355.

The Decay of ^{188}Re

By R. RICHMOND, P. J. GRANT* and H. ROSE
Cavendish Laboratory, University of Cambridge

Communicated by E. S. Shire; *MS. received* 28*th August* 1951, *and in amended form* 3*rd March* 1952

ABSTRACT. The gamma-rays emitted in the decay of ^{188}Re have been investigated by means of absorption and coincidence techniques and by the use of a thin lens beta-ray spectrometer. The gamma-ray intensities (expressed as a percentage of the total number of beta-disintegrations) have been measured and a probable decay scheme is proposed.

§1. INTRODUCTION

Slow neutron irradiation of rhenium produces the two radioactive isotopes ^{186}Re and ^{188}Re which decay by beta-emission to the osmium isotopes ^{186}Os and ^{188}Os with half-lives of 90 hours and 18 hours respectively. ^{188}Re has previously been investigated by Miller and Curtiss (1946), Goodman and Pool (1947), Cork, Shreffler and Fowler (1948) and Beach, Peacock and Wilkinson (1949), but the only gamma-ray intensity measurements have been those of Miller and Curtiss who gave rough estimates of the relative intensities of the gamma-rays.

---

* Now at the Department of Natural Philosophy, Glasgow University.

§ 2. THE DETERMINATION OF GAMMA-RAY INTENSITIES

The secondary electron spectrum ejected by the gamma-rays from a thick lead radiator (150 mg cm^{-2}) was measured in the spectrometer and is shown in fig. 1. Photoelectron peaks corresponding to gamma-rays of energies $0{\cdot}152 \pm 0{\cdot}001$ Mev (K and L levels), $0{\cdot}476 \pm 0{\cdot}005$ Mev (K only), $0{\cdot}638 \pm 0{\cdot}005$ kev (K), and $0{\cdot}933 \pm 0{\cdot}008$ Mev (K) were observed (the 0·152 Mev peak is not shown in the figure). These values are in good agreement with those obtained by Miller and Curtiss and by Beach *et al.* The 1·40 Mev gamma-ray reported by these authors was too weak to be detected in the present experiment.

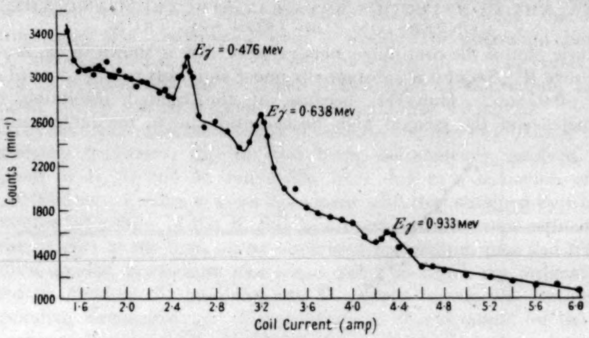


Fig. 1. Photoelectron spectrum of ^{188}Re γ-rays.

Using the method of Ellis and Aston (1930) as modified by Richardson (1950) the relative intensities of the 0·476, 0·638 and 0·933 Mev gamma-rays were estimated from the secondary electron spectrum to be in the ratio 1 : 2 : 1·5 with an accuracy within about 10%. Since the loss of electrons from the peak of the line by multiple scattering and anisotropic emission of the photoelectrons may be very serious at low energies, no attempt was made to compare the intensities of the hard gamma-rays with that of the 0·152 Mev radiation.

The absolute values of the gamma-ray intensities were determined by taking an absorption curve of the gamma-radiation in lead, using a Geiger counter whose efficiency as a function of gamma-ray energy had previously been estimated. The curve could be resolved into four components corresponding to gamma-rays of energies 0·15, 0·42, 0·96 and 1·32 Mev. These values are in satisfactory agreement with those obtained on the spectrometer. No component corresponding to the 0·638 Mev gamma-ray could be resolved and it was assumed that the '0·42 Mev' component contains the 0·476 Mev and the 0·638 Mev gamma-rays since these have similar mass absorption coefficients. The relative intensities of the four resolved components were determined from the absorption curve, and the absolute intensities, expressed as a percentage of the total number of beta-disintegrations, were then estimated using a calibrated beta-counter in conjunction with the gamma-counter. The results are:

Gamma-ray energy (Mev)	0·15	0·42	0·96	1·32
Gamma-ray intensity (Total β-ray intensity = 100)	70 ± 10	7 ± 2	5 ± 2	5 ± 2

These figures are of low accuracy because the continued subtraction of the higher energy components from the absorption curve leads to cumulative errors. They are combined with the more accurate relative intensity values obtained by means of the beta-ray spectrometer to give the following estimate of the absolute intensities of all the gamma-transitions:

Gamma-ray energy (Mev)	0·152	0·476	0·638	0·933	1·40
Gamma-ray intensity	70 ± 10	3 ± 1	6 ± 2	4·5 ± 2	4·5 ± 2
(Total β-ray intensity = 100)					

§ 3. THE CONTINUOUS BETA-RAY SPECTRUM AND THE CONVERSION ELECTRON SPECTRUM

A Kurie plot of the continuous beta-ray spectrum is shown in fig. 2. In the region above $W = 3m_0\mathbf{c}^2$ it is substantially linear and leads to an end point energy of $2{\cdot}07 \pm 0{\cdot}02$ Mev. However, because of the limited resolution of the spectrometer and the general high background due to the large gamma-ray intensity it is not certain whether this end-point energy refers to the transition to the ground state, the transition to the 0·152 Mev level in ^{188}Os or to some mean of these two transitions. This difficulty does not affect the decay scheme put forward in § 5, but does give an uncertainty to the exact disintegration energy (shown as the dotted line at ~2·1 Mev in fig. 5).

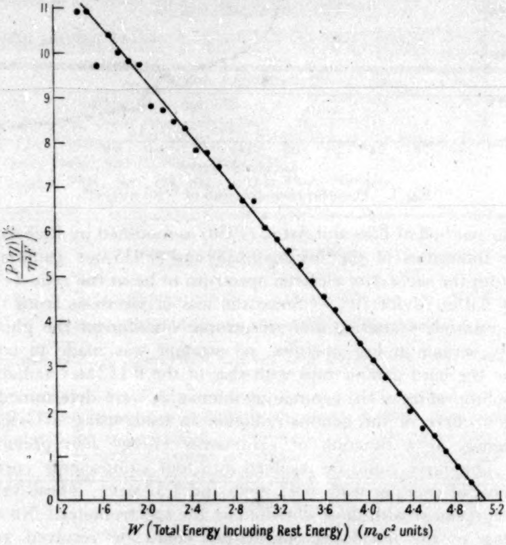


Fig. 2. Kurie plot of ^{188}Re continuous β-ray spectrum.

Below $W = 3m_0\mathbf{c}^2$ the beta-spectrum of ^{188}Re was obtained by subtracting the contribution of the ^{186}Re spectrum (which was measured after the ^{188}Re had been allowed to decay for a period of 10 half-lives), and is therefore subject to cumulative errors. The Kurie plot in this region is still substantially linear, but this cannot be taken to indicate that no beta-ray branching occurs since the plot is not of sufficient accuracy to show a branching of less than about 10%. In fact, gamma-ray intensity measurements and β–γ coincidence experiments (see § 4 below) show that such branching is present.

The conversion electron spectrum was investigated in an attempt to determine the multipolarity of the 0·152 MeV gamma-ray. The spectrometer resolution ($\Delta\eta/\eta$ at half-maximum of a conversion line, where η = electron momentum) was 5%. The Geiger counter used for detecting the electrons had a thin nylon window which transmitted electrons with energies greater than 10 kev. Rhenium metal was precipitated from a solution of potassium perrhenate ($KReO_4$) and deposited as a source of thickness 0·5 mg cm^{-2}. The source backing consisted of a nylon film on to which a thin layer of aluminium had been evaporated, giving a total thickness of approximately 0·2 mg cm^{-2}.

Three conversion lines were found at electron energies of 0·0779, 0·1388 and approximately 0·150 MeV, the last two being incompletely resolved. These correspond to K, L and M conversion lines due to a transition of energy 0·152 ± 0·0005 MeV, a value in good agreement with that obtained by Cork *et al.* The relative intensities of the K and L conversion lines were estimated both from the heights of the lines on an undivided momentum plot and from their areas on a divided momentum plot (i.e. a graph in which the ordinate $P(\eta)$ is obtained by dividing the counting rate at a given momentum setting by the corresponding momentum η). The mean value thus obtained for the ratio of the number of K conversion electrons to the number of L conversion electrons N_K/N_L was 0·42 ± 0·04. By comparing areas on the divided momentum plot the ratio of L conversion electrons to beta particles N_L/N_β was found to be 0·076 ± 0·008. By the same method a value of 0·032 ± 0·004 was obtained for the ratio N_K/N_β and from this value the ratio of K-conversion electrons to gamma-quanta N_K/N_Q was estimated to be 0·046 ± 0·01.

An empirical curve of the ratio N_K/N_L for electric quadrupole radiation by Goldhaber and Sunyar (1951) predicts a value of 0·5–0·6 for ^{188}Os which agrees well with the observed value. On the other hand according to the tables of Rose *et al.* (1951) the experimental value of N_K/N_Q agrees best with the calculated value for electric dipole radiation and is less than the value for electric quadrupole radiation by a factor of 7:

	Electric			Magnetic			Observed
l	1	2	3	1	2	3	
N_K/N_Q	0·11	0·33	0·93	1·58	8·00	29·0	0·046 ± 0·01

It does not seem reasonable to attribute this discrepancy to experimental error in the determination of the intensity of the 0·152 MeV gamma-ray because intensity measurements of the 0·136 MeV gamma-ray of ^{186}Re, using the same experimental equipment, have given a value agreeing well with the intensity of the associated partial beta-spectrum measured on the spectrometer (Grant and Richmond 1949; see also Steffen 1951). Scattering and absorption due to non-uniformity in the source undoubtedly reduced the number of K-conversion electrons observed, but careful calculation shows that this effect could not have reduced the K-conversion peak by more than about 15% at most. It would therefore appear that there is a real and substantial disagreement between the theoretically predicted number of K-conversion electrons and the number actually emitted.

§4. COINCIDENCE MEASUREMENTS

Curves of the absorption in lead of γ–γ coincidences are shown in figs. 3 and 4. These were taken using two photomultipliers (E.M.I. type 6262) in conjunction with a coincidence circuit of resolving time 2×10^{-8} second. The curve of fig. 3 was obtained by placing equal thicknesses of lead absorber between the source and each multiplier and measuring the coincidence counting rate as a function of absorber thickness. Curve A of fig. 4 was obtained by placing different thicknesses of lead absorber between the source and one multiplier only. A lead absorber of thickness 1·8 g cm^{-2} was then placed between the source and the other multiplier and the experiment repeated to give curve B. In all cases aluminium absorbers were placed adjacent to the source to absorb the beta-particles.

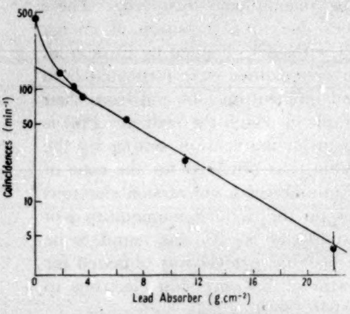


Fig. 3. Absorption of γ–γ coincidences (absorbers on both sides of source).

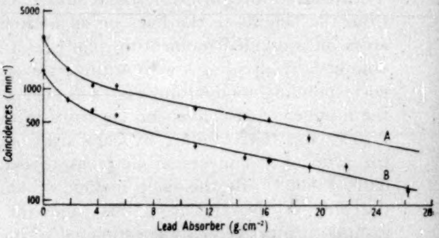


Fig. 4. Absorption of γ–γ coincidences (absorbers on one side of source).

Observations of β–γ coincidences were also made, the beta-particles being excluded from one multiplier by means of an aluminium absorber. Gamma-ray absorption measurements showed that beta-particles were in coincidence with the 0·152 Mev gamma-ray and also with the harder gamma-rays. Absorption of the beta-particles in aluminium showed that particles of energies greater than 1·5 Mev were in coincidence only with the 0·152 Mev gamma-ray.

Coincidence measurements made after the decay of the ^{188}Re showed that ^{186}Re radiations could not have affected the above results.

§5. DECAY SCHEME

The five gamma-rays may most simply be fitted to the decay scheme shown in fig. 5. The deductions which may be made from the experimental results are summarized below and it will be seen that they are all in accordance with this scheme.

Intensity Measurements

(i) The 0·152 Mev gamma-ray has an intensity four times as great as the total intensity of the four harder gamma-rays, and must therefore come from the lowest excited state. (ii) Since the maximum beta-ray energy is about 2·1 Mev, energy considerations show that the 1·40 MeV and 0·933 MeV gamma-rays cannot be in coincidence. Coincidences between the 1·40 MeV gamma-ray and the 0·638 and 0·476 MeV gamma-rays would correspond to beta-transitions of energies about 0·1 and 0·2 MeV respectively. In either case the intensity of the 1·40 MeV gamma-ray would be very much less than the observed value of 4·5%. The 1·40 MeV gamma-ray can therefore be in coincidence only with the 0·152 MeV gamma-ray.

Coincidence Measurements

In the coincidence absorption curves only three components may be differentiated: (*a*) the 0·152 MeV gamma-ray; (*b*) a component containing either the 0·476 MeV or the 0·638 MeV gamma-ray or both; (*c*) a component containing either the 0·933 MeV or the 1·40 MeV gamma-ray or both.

(i) The curve of fig. 3 shows only components (*a*) and (*b*). This is in agreement with the proposed decay scheme since, when the 0·152 MeV gamma-ray has been removed, only coincidences between the 0·933 MeV gamma-ray and the two gamma-rays of component (*b*) will be recorded and each of these coincidences will have the same absorption coefficient as the lower energy member of component (*b*). (ii) The curve A of fig. 4 can be resolved to show all three components. Therefore all three components are involved in coincidences. (iii) In curve B of fig. 4 the 1·8 g cm^{-2} absorber reduces the 0·152 MeV gamma-ray to 4% of its original intensity without appreciably affecting the other gamma-rays. Hence this curve shows the presence of coincidences between the 0·933 MeV gamma-ray and either the 0·476 MeV or the 0·638 MeV gamma-ray or both. (iv) The parallelism of curves A and B shows that the 0·152 MeV gamma-ray is in coincidence with both (*b*) and (*c*).

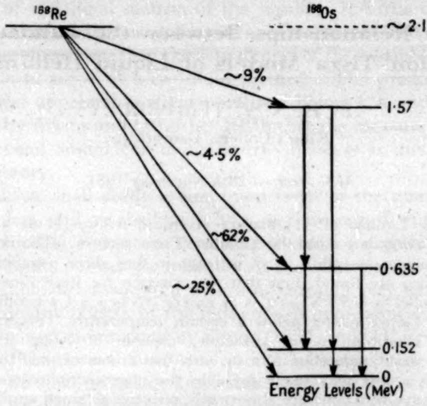


Fig. 5. Decay scheme of ^{188}Re.

While none of the experimental evidence contradicts the proposed scheme, the ambiguity attached to components (*b*) and (*c*) makes it possible to construct more complicated schemes (containing five energy levels) which would give the same experimental results.

ACKNOWLEDGMENTS

Our thanks are due to members of the Atomic Energy Research Establishment, Harwell, where neutron irradiation of the rhenium samples was carried out, and also to Dr. A. G. Maddock for assistance in the preparation of the sources. We are indebted to Mr. J. M. C. Scott for helpful discussions.

REFERENCES

Beach, L. A., Peacock, C. L., and Wilkinson, R. G., 1949, *Phys. Rev.*, **76**, 1585.
Cork, J. M., Shreffler, R. G., and Fowler, C. M., 1948, *Phys. Rev.*, **74**, 1657.
Ellis, C. D., and Aston, G. H., 1930, *Proc. Roy. Soc.* A, **129**, 180.
Goldhaber, M., and Sunyar, A. W., 1951, *Phys. Rev.*, **83**, 906.
Goodman, L. J., and Pool, M. L., 1947, *Phys. Rev.*, **71**, 288.
Grant, P. J., and Richmond, R., 1949, *Proc. Phys. Soc.* A, **62**, 573.
Miller, L. C., and Curtiss, L. F., 1946, *Phys. Rev.*, **70**, 983.
Richardson, H. O. W., 1950, *Proc. Phys. Soc.* A, **63**, 234.
Rose, M. E., Goertzel, G., Spinrad, B., Harr, J., and Strong, P., 1951, *Phys. Rev.*, **83**, 79.
Steffen, R., 1951, *Phys. Rev.*, **82**, 827.

On the Relationships Between the Landau and London–Tisza Models of Liquid Helium II

By H. N. V. TEMPERLEY
King's College, Cambridge

MS. received 28*th November* 1951

ABSTRACT. The Landau (1941) picture of liquid helium II as a modified Debye model is critically compared with the London–Tisza picture of liquid helium II as a condensed Bose–Einstein assembly in an attempt to find some common features. Two such common features are found, first that the criterion for Bose–Einstein condensation in a one-particle model is $(\partial S/\partial N)_{E,V} \leqslant 0$ or $g=(\partial F/\partial N)_{V,T} \geqslant 0$, a condition that can also be satisfied by the Debye model below a certain temperature. Secondly, an assembly satisfying such a condition, is in principle, capable of being in thermodynamic equilibrium with a weakly adsorbed film or with free atoms of small energy. It is also suggested that such atoms may make possible the observed phenomena of superfluid flow because their wave-functions are abnormally sensitive to small applied external fields of force, and some evidence in support of this is presented.

A numerical study of the Landau–Debye model shows that, in accordance with observed facts on helium II, its energy spectrum is probably modified considerably in channels of less than 10^{-4} cm width. It is also concluded that the Debye model is probably not a workable approximation near the λ-point, but that it is correct at sufficiently low temperatures.

§1. INTRODUCTION

The behaviour of liquid helium can be quantitatively described by means of the 'two-fluid' picture, which is the assumption that liquid helium behaves as a solution of two fluids, each with its own density and stream velocity, one of them, the superfluid, being devoid of viscosity and unable to carry any entropy, while the other component behaves like an ordinary liquid, with a finite viscosity that can be measured by an oscillating disc type of experiment (Andronikashvili 1948). It is also necessary to postulate that the 'superfluid' effects disappear progressively as the velocities increase, and Gorter and Mellink (1949) have shown that many of the experimental facts can be interpreted by the assumption of a mutual friction between the two fluids and there is also evidence of a critical velocity of superfluid flow (Kapitza 1941, Atkins 1951, Bowers and Mendelssohn 1950). Gorter and Mellink also find some evidence of a decrease in the effective viscosity of the normal fluid for very narrow channels ($\sim 10^{-4}$ cm) which they suggest may be a mean free path effect. Also, for channels narrower than this approximate width, the critical velocity of superfluid flow begins to increase with diminishing width of channel, this critical velocity being roughly inversely proportional to the width.

Two physical models which lead to the two-fluid theory as a consequence have been proposed. F. London (1939) has suggested that the perfect gas model is a satisfactory first approximation, and that the appearance of superfluidity is connected with the condensation phenomenon that occurs in a perfect gas obeying Bose–Einstein statistics. Landau (1941) takes the Debye model of a solid as a first approximation, supplementing it by postulating the presence of 'rotons' as well as ordinary sound quanta or 'phonons'. The 'roton' is supposed to be a quantized state of rotational motion of the liquid. It turns out that the total effective mass of these excitations, defined in a special way to be described below, is, at very low temperatures, less than the actual mass of the assembly, and in this way it is also possible to arrive at a two-fluid picture. The predictions of the two theories only differ appreciably at temperatures below 1° K and in this region it has been shown by Atkins and Osborne (1950) that the measured velocity of propagation of 'second sound' or temperature waves is in much closer accord with Landau's theory.

In this paper we shall study a simplified form of the quantum-mechanical many-body problem, and we shall show how, as the strength of the interactions is progressively reduced, we pass over continuously from a slightly modified form of Landau's model to the perfect gas model. At the same time, interpretations will be given of the anomalies associated with flow through very narrow slits noticed by Gorter and Mellink (1949), of the influence of zero-point energy on the properties of liquid helium, pointed out by Simon (1934) and of the peculiar behaviour of the viscosity of the normal fluid as a function of temperature (Andronikashvili 1948). Finally, we shall point out a possible reason for the occurrence of superfluidity or, more precisely, flow of liquid helium without appreciable transport of entropy under very small pressure heads.

§ 2. THE RELATIVE SUCCESSES OF THE TWO MODELS

We now give a list of the experimental facts (*a*) that do not point decisively in either direction, (*b*) that seem to confirm the Landau type model, (*c*) that seem to confirm the Bose–Einstein type model. Under heading (*a*) we may first of all mention the equilibrium properties such as the equation of state and the specific heat curve. Neither of these show anything particularly unusual except for the small negative expansion coefficient and absence of a triple point. The anomaly in specific heat is of a type very commonly encountered. As Simon (1934) has pointed out, there is evidence of an abnormally high zero-point energy (of the order of 70 cal/mole) which has the effect of making the lattice spacing abnormally high. A theory of Debye type gives a zero-point energy of $9N\boldsymbol{k}\Theta/8$ which is of the right order of magnitude. Temperley (1947) has pointed out that a generalization of the perfect gas model, in which each atom is supposed to be moving in a spatially periodic field representing its interactions with the other atoms, also possesses a zero-point energy and may have a coefficient of expansion of either sign.

A second set of experimental facts under this heading seems to be those that can be interpreted by the two-fluid theory, without enabling one to calculate a definite value of the ratio ρ_s/ρ_n. Among these we may mention the thermo-mechanical effect, the experiments on heat transport, discussed by Gorter and Mellink (1949) and the experiments on flow in capillaries discussed by Atkins (1951).

A third set of experimental facts, the oscillating disc type of experiment and measurements of the velocity c_2 of second sound above 1° K do lead to definite values of the ratio ρ_s/ρ_n (Gorter, Mellink and Castelejn 1950) but the required values of this ratio are consistent with either theory.

Under heading (*b*) come the experiments of Atkins and Osborne (1950) on the velocity c_2 of second sound below 1° K. These confirm entirely the results of earlier work which indicated a rapid rise in c_2 below 1° K and point to values of c_2 in close agreement with Landau's theory. As has been shown (Temperley 1951 a, Dingle 1951), such results can only be obtained from a theory of London–Tisza type if one abandons the implicit assumption that $\rho_s/\rho = N_0/N$ where N_0 is the occupation of the lowest state. A second piece of evidence is the steep rise in the viscosity of the normal fluid with falling temperature in this region, which is, as we shall see, characteristic of a 'solid-like' rather than a 'gas-like' model.

Under heading (*c*) comes the occurrence of a transition point. According to Landau (1941) the transition to HeI should occur when the effective density ρ_n as defined by him becomes equal to the actual density ρ, but his theory in its present form gives no explanation at all of why this equalization should be accompanied by a co-operative transition. Such a transition is, however, a natural consequence of the Bose–Einstein type of theory (F. London 1939). A second piece of evidence that a 'gas-like' model may be more appropriate near the transition point is the fact that the viscosity is rising with increasing temperature in this region (as well as for He I).

What might seem to be a third piece of evidence is that the formation of thick and mobile adsorbed films below the transition temperature receives a natural explanation (Temperley 1949, Band 1951), if one supposes that the adsorbed film is in equilibrium with a condensed Bose–Einstein assembly. The Landau theory, in its present form, gives no such explanation, but we shall see that if properly developed the Landau theory could explain this fact; thus this piece of evidence really belongs in category (*a*).

Taken all together, the evidence appears to indicate that the Debye-type model is a good first approximation at very low temperatures, but that a 'gas-like' model may be more appropriate near the transition point. The behaviour of the viscosity of the normal fluid, which apparently passes through a flat minimum at about 1·7° K, also suggests that more than one mechanism is responsible for it.

The Debye model will now be examined quantitatively and it will be shown that it probably is a good approximation at very low temperatures. The manner in which it passes over into a 'gas-like' model as the interactions are weakened or as the temperature rises will also be traced. A study of its entropy, as a function of the number of atoms present, shows that the model is theoretically capable of a 'condensation effect' which has not been noted before. This seems to provide a connecting link between the two types of model, and to give another instance of the already known fact, see §5, that the Bose–Einstein condensation phenomenon is not confined to 'one-particle' models but that analogues can exist in the many-body model even with very strong interactions. On this basis, the formation of thick adsorbed films can be understood on the basis of the Landau model (1941) as well as on the London–Tisza model (F. London 1939).

§ 3. THE DEBYE TYPE MODEL

It is known (see, for example, Peierls 1929) that the Debye model is practically equivalent to the assumption that the potential energy of a crystal lattice is a quadratic function of the distances of the atoms from their equilibrium positions, or from one another. Peierls uses the linear lattice to illustrate the departure of the modes of an actual atomic lattice from the ideal Debye spectrum. We may write $V=\frac{1}{2}m\omega_0^2\,\Sigma_r(x_r-x_{r+1})^2$ and obtain for the classical equations of motion

$$\frac{d^2x_r}{dt^2}+\omega_0^2(2x_r-x_{r-1}-x_{r+1})=0. \qquad \ldots\ldots(1)$$

By making the substitution $\xi_s=\Sigma_r x_r\exp(2rs\pi i/N)$ Peierls finds for the possible modes of oscillation

$$\omega_s=2\omega_0\sin(s\pi/N). \qquad \ldots\ldots(2)$$

These differ from the ideal Debye spectrum in that the possible frequencies are proportional to $\sin(s\pi/N)$ instead of to s, which means that one automatically gets N normal modes without having to assume an arbitrary cut-off of the spectrum as Debye does. If we write the quantum-mechanical version of eqn. (1) we get

$$\frac{\boldsymbol{h}^2}{8\pi^2m}\sum_r\frac{\partial^2\psi}{\partial x_r^2}+E\psi-\frac{m\omega_0^2}{2}\sum_r(x_r-x_{r+1})^2\psi=0. \qquad \ldots\ldots(3)$$

To avoid end effects, we may suppose the atoms arranged in a ring of total length l and that the last atom interacts with the first, the interaction between them being proportional to $(x_1-x_N+l)^2$ rather than to $(x_1-x_N)^2$. This secures that the total energy is a minimum when the atoms are spaced equally round the ring, instead of when they are all bunched together. Symmetry between the N atoms can then be restored if we write $y_1=x_1, y_2=x_2-l/N, y_3=x_3-2l/N\ldots$ etc. This makes the potential energy again a quadratic function of the coordinates, and the terms involving $\omega_0^2l^2$ simply imply a change of the energy zero. Equation (3) becomes separable if we write $C_s=\Sigma_r y_r\cos rs\theta$, $S_s=\Sigma_r y_r\sin rs\theta$ (where $\theta=2\pi/N$). It becomes

$$\left\{\frac{N\boldsymbol{h}^2}{8\pi^2m}\sum_s\left(\frac{\partial^2}{\partial C_s^2}+\frac{\partial^2}{\partial S_s^2}\right)+E-V_0-\frac{2\boldsymbol{m}\omega_0^2}{N}\sum_s\sin^2\left(\frac{s\theta}{2}\right)(C_s^2+S_s^2)\right\}\psi=0. \qquad \ldots\ldots(4)$$

A typical solution of eqn. (4) is

$$\psi=\prod_s\exp\left(-\frac{C_s^2+S_s^2}{2r_s^2}\right)H_t\left(\frac{C_s}{r_s}\right)H_u\left(\frac{S_s}{r_s}\right), \qquad \ldots\ldots(5)$$

where the H_t, H_u are Hermite polynomials, not the same for all values of s, the order of each of these polynomials indicating the number of sound quanta or 'phonons' associated with the corresponding variable. The energy associated with the variable C_s or S_s is a half-odd multiple of $\hbar\omega_s$, where ω_s is given by eqn. (2), and r_s is a distance given by $(N\boldsymbol{h}/4\pi m\omega_0\sin\frac{1}{2}s\theta)^{1/2}$, and is a measure of the range through which C_s or S_s can depart from the zero values corresponding to uniform spacing of atoms round the ring before the wave function ψ becomes negligible.

We can obtain a relation between the constant ω_0 specifying the strength of the interactions and the measured velocity of sound c_1 at low frequencies by an application of the correspondence principle. As Peierls (1929) remarks, in a problem of this kind the wavelength cannot be strictly defined, and its place is taken by the parameter s, which specifies the phase difference between neighbouring atoms associated with phonons involving the variable C_s or S_s. For small values of s this phase difference is practically the same as that associated with a sound of wavelength l/s, so that we may deduce the relation between c_1 and ω_0

$$c_1 = \lambda\nu = \frac{l}{s}\,\frac{2\omega_0}{2\pi}\sin\left(\frac{s\pi}{N}\right) \simeq \frac{l\omega_0}{N}. \qquad \ldots\ldots(6)$$

If we wish to preserve the acoustic concept of a wavelength l/s for large values of s, it is necessary to admit that the corresponding velocity of sound depends on s. The changes necessary if we work with an atomic lattice instead of a continuum are an upper limit to the possible frequencies, given according to eqn. (2) by ω_0/π, and a dispersion in the velocity of sound. Substituting for ω_0 in the relation for r_s we get

$$r_s \simeq (N\boldsymbol{h}l/4\pi^2 mc_1 s)^{1/2} \qquad \ldots\ldots(7)$$

which enables us to estimate the distance y_r by which a single molecule is shifted from its equilibrium position as a result of the zero-point motion associated with a quantized sound wave of wavelength l/s. For small values of s, the corresponding classical sound wave would be represented by a space variation of the type $y_r = y_0 \cos(rs\theta)$, y_0 being the amplitude. Comparing this with the expression $C_s = \Sigma_r y_r \cos(rs\theta)$ and the condition that C_s may attain values of the order of r_s, we conclude that

$$y_0 \simeq 2r_s/N \simeq (\boldsymbol{h}l/N\pi^2 mc_1 s)^{1/2}. \qquad \ldots\ldots(8)$$

A result very similar to (8) follows if we suppose that the energy $\frac{1}{2}\boldsymbol{h}\nu_s$ is shared among N molecules. Taking the case $s=1$ and inserting numerical values $l/N = 3{\cdot}6 \times 10^{-8}$ cm, $c = 2{\cdot}5 \times 10^4$ cm/sec and $m = 6 \times 10^{-24}$ g appropriate for liquid helium, we find $y_0 \sim 10^{-8}$ cm; that is to say that the zero-point motion has amplitudes comparable with the atomic spacing, a result that has been pointed out by several writers.

§ 4. THE LIMITATIONS OF THE DEBYE MODEL

We have to consider first the changes that have to be made in the one-dimensional Debye model to make it apply to a three-dimensional lattice, secondly the manner in which it is likely to break down as the interactions are reduced or the temperature raised, thirdly the modifications necessary to allow for the symmetry properties of the helium atoms, and lastly the effects of cubic and higher terms in the energy.

(i) *The Change to the Three-Dimensional Case*

This question has been considered by Peierls (1929) and, in more detail, by Blackman (1935). As Peierls shows, the introduction of interactions between other than nearest neighbours in the linear model introduces no new feature of physical interest while, in the two and three dimensional cases, the introduction of 'cross-bracing' interactions between second nearest neighbours implies the possibility of propagation of transverse or shear waves. Temperley (1951 b) has suggested that Landau's 'rotons' might well be replaced by quantized shear waves which do represent rotational motion, and that the available evidence is consistent with their velocity of propagation being comparable with, perhaps even somewhat greater than, the ordinary velocity of sound. The introduction of these extra interactions makes the energy spectrum more complicated but as long as we confine ourselves to terms in the interaction energy V that are quadratic in the coordinates, the equation can still be separated. One can always find the orthogonal transformation of coordinates that reduces such a function V to a sum of squares, and such a transformation changes $\Sigma_r \Delta_r^2$ into the same operator in the new coordinates. Associated with each of these coordinates, we can define an 'effective velocity of sound' by a process analogous to that leading to eqn. (6). The correspondence principle shows that the continuum treatment must be accurate at sufficiently low frequencies but even here we may have to allow for both longitudinal and transverse oscillations. We thus arrive at the ordinary Debye model with zero-point energy $\frac{1}{2}\hbar\omega_s$ associated with each normal coordinate, the longitudinal and transverse velocities of sound being in general different, and both varying with frequency. The dispersion is a consequence of the fact that we are dealing with a discontinuous lattice, and is already present in the one-dimensional model.

The physical picture of a phonon arrived at above differs in some respects from the rather vague picture of a 'wave packet of sound' (localized in space and travelling with the group velocity), that most writers seem to have in mind. Our assembly is to be described by a superposition of states $\psi(t_s, u_s \dots)$ given by (5), where the t's and u's take all possible values, but, in practice, a complexion in which t_s departs appreciably from its statistical mean value $1/\{\exp(\hbar\omega_s/\boldsymbol{k}T)-1\}$ is very unlikely, so that the assembly is probably described sufficiently accurately by a *pure* wave function in which the t's and u's have values near to these mean values. If so, the emission or absorption of a phonon of frequency $\omega_s/2\pi$ corresponds simply to a change in t_s or u_s of unity, but such a process affects *all the atoms on an equal footing*, and cannot therefore be associated with any definite part of the assembly. We cannot meet this difficulty by describing the assembly by a mixed rather than by a pure wave function without blurring the concept of emission or absorption of a phonon (we cannot get a clear wave-mechanical picture of the absorption of a quantum of radiation, unless we begin with an atom in a definitely specified state). We can, however, use Peierls' device (1929) of dividing up our assembly into L sub-assemblies of N/L atoms, and then assign a set of wave functions to each sub-assembly. The disappearance of a phonon from one sub-assembly and its appearance in another can then be given a definite meaning, but it remains true that each phonon must be associated with a large number of atoms and therefore that it cannot be precisely localized.

.A
5
2

Another important point is that the energies and frequencies of phonons are quite unaffected by a uniform external field such as gravity. For the addition of a potential energy $mg(y_1+y_2+\dots)$ to eqn. (4) affects only the equation involving $C_0=(y_1+y_2+\dots)$ which governs the motion of the centre of gravity of the assembly, and none of the equations involving C_s or S_s $(s \neq 0)$. (A non-uniform field, involving something like $\Sigma_r y_r^n$ would introduce stresses into the assembly, and this could, of course, affect the frequency spectrum.) We can also show that the energy of the phonons is unaffected by a Galilean transformation of coordinates. For, if we put $y_r' = y_r + vt$, $t' = t$, we have $\partial/\partial y_r' \equiv \partial/\partial y_r$ $\partial/\partial t \equiv \partial/\partial t' + v\Sigma_r \partial/\partial y_r'$ so that the change in energy of the whole assembly$(\boldsymbol{h}/2\pi i)(\partial/\partial t' - \partial/\partial t)$ is $(v\boldsymbol{h}/2\pi i)\Sigma_r \partial/\partial y_r' = (v\boldsymbol{h}/2\pi i)\partial/\partial C_0$, which energy change is all attributable to the change in velocity of the centre of gravity of all the atoms, and there is *no* energy change associated with the phonons themselves. Wave functions involving C_s or S_s actually correspond classically to stationary sound waves, but the above result remains true even if we use the quantum analogue of a travelling sound wave (see for example Wilson 1936, chap. 6). Landau's (1941) derivation of the effective mass of an assembly (see p. 503) seems to be based on the assumption that phonons behave like material particles and that the energy of *each one* is changed by pV by a Galilean transformation of axes. However the change in energy of any assembly of material particles, such as a Debye lattice, must, as we have seen, be proportional to their total momentum, that is, to the momentum of the centre of gravity, so that Landau's derivation needs revision. Nevertheless, it is possible to obtain results, precisely equivalent to Landau's for the effective mass of an assembly by a somewhat different argument, which will be found in the Appendix. In this revised derivation the 'momentum of a phonon' is defined simply as the momentum that would be imparted to a material particle if the phonon disappeared, and nothing is assumed about any change of energy of an individual phonon as a result of a Galilean transformation.

(ii) *The Effect of Raising the Temperature*

At extremely low temperatures, the energy associated with the normal coordinate C_s is $\frac{1}{2}\hbar\omega_s$ where ω_s is given by eqn. (2) and, as we have seen, the corresponding migration of an atom from its equilibrium position associated with one particular frequency is of the order of 10^{-8} cm. If, however, we raise the temperature, the probable energy associated with C_s is $\hbar\omega_s/\{\exp(\hbar\omega_s/\boldsymbol{k}T)-1\}\simeq\boldsymbol{k}T$ for $\boldsymbol{k}T\gg\hbar\omega_s$ and the corresponding migration of an atom is multiplied by a factor of the order of $(\boldsymbol{k}T/\hbar\omega_s)^{1/2}$. This factor will be largest when $s=1$ and, as we have seen, this value of s already gives the largest migrations at absolute zero. Thus, according to the Debye theory, at 1° K the thermal vibrations of the lattice are already displacing individual atoms through distances of the order of $(\boldsymbol{k}T/\hbar\omega_s)^{1/2}\times10^{-8}\simeq10^{-5}$ cm, and the conception of liquid helium as a crystal lattice with the atoms bound in the neighbourhood of equilibrium points is apparently becoming far-fetched. Nevertheless, at least one other instance is known of a bound quantum-mechanical system in which both the potential and kinetic energies are large compared with the actual binding energy, namely the deuteron. The characteristic feature of this system is that the proton and neutron are very likely to be at a distance apart which is significantly larger than the range of action of the force between them, but nevertheless they continue to form a bound state. Furthermore, the behaviour of such a system is not very sensitive to the precise form of the interaction as a function of the distance, but only to various weighted averages of this function. In an analogous way, it seems possible to understand why in liquid helium the crude assumption of a square law form of interaction (which is certainly not valid for an atom that has migrated as much as one lattice distance) can nevertheless give results in reasonable agreement with experiment.

To follow the changes that take place as the temperature rises or as the interactions are weakened, we should have to assume somewhat different boundary conditions in eqn. (4). So far, we have implicitly assumed those appropriate to the ordinary simple harmonic oscillator problem, namely that the wave functions must represent bound states, and must thus vanish sufficiently fast as any of the $C_s\to\infty$. If the interactions were weaker, we should have to take account of the effect of the walls, and the problem would become similar to that of simple harmonic oscillators enclosed between rigid walls, which has been treated by Auluck and Kothari (1945). The effect on the energy spectrum would become noticeable at 'ordinary' helium temperatures when the walls were at distances of the order of 10^{-5} cm which may be the explanation of the fact, noted by Gorter and Mellink (1949), that the calculated viscosity of the normal fluid apparently falls off sharply for slits of and below this width, and of the increase in the critical velocity of superfluid flow for such slits. The calculated figure of 10^{-5} cm for the migration of a typical atom associated with non-zero values of C_1 or S_1 seems at first sight rather small, both the above effects already being quite noticeable for slits of 10^{-4} cm width. We must remember, however, that this calculated figure represents the migration of an atom associated with one frequency only, whereas there will be a displacement of an atom associated with each of the normal modes. These displacements have to be added up in a random manner because, in a solution such as (5), the phases of C_s and S_s are unspecified. In this way it seems easily possible to account for the discrepancy of a factor of 10 or so.

(iii) *The Symmetry Properties of the Atoms*

So far we have taken no account of the fact that our theory is to be invariant if we interchange one or more pairs of atoms. The interaction energy in an equation such as (3) ought to be a symmetrical function of the coordinates, whereas we have neglected all interactions except those between nearest neighbours. This could be formally remedied by replacing the interaction function by one such as $\Sigma_r\Sigma_s V_2(x_r - x_s)^2$ (which would actually lead to a model of Einstein rather than Debye type, all the normal modes having the same frequency). Such a model, however, implies that the interaction between distant atoms is far greater than that between nearest neighbours, and it is, for that reason, unphysical. What we really require, to represent a solid, is an interaction energy that is always large except for configurations corresponding to a nearly perfect crystal lattice.

For such a model, it is shown by Rushbrooke (1949) that the indistinguishability of the atoms does not matter, and that we can get the correct partition function even if we make the physically inadmissible assumption that the atoms are arranged on the lattice in a fixed numerical order. In quantum-mechanical terms, we obtain the same set of energy levels whether we use wave functions such as (5), or the correct ones obtained from these by the symmetrizing process of permuting the atoms and superposing the resulting wave functions. As we weaken the interactions, exchanges of places between the atoms become more and more frequent, and it becomes a poorer approximation to use wave functions such as (5). Finally, when the interactions become very weak, we are back to the perfect gas model, for which the symmetry properties of the atoms are very important. Thus, we reach Wergeland's conclusion (1947) that we have to consider the symmetry properties of the atoms when, and only when, the interactions are weak.

(iv) *The Effect of the Cubic Terms in the Energy*

Peierls (1929) showed that, even in an ideal lattice at very low temperatures, it is quite essential to consider the effect of the cubic terms in the interaction energy, otherwise the thermal conductivity of the lattice diverges. This remains true even if we postulate the existence of regions in the lattice where the effective value of the velocity of sound is different from that in the surrounding regions. Scattering proportional to ν^4 occurs, but the thermal conductivity still diverges because of an 'infra-red catastrophe'. A similar result is easily seen to be true for viscosity. Taking both the irregularities and the cubic terms into account, Peierls (1929) considered three-phonon 'Umklapp' processes and concluded that the conductivity at very low temperatures should behave like $1/T$. Landau and Khalatnikov (1949) considered four-phonon processes without 'Umklapp' and concluded that the viscosity due to phonons only should behave like T^{-5}, and they also found it essential to take dispersion into account.

This type of variation of the transport coefficients with temperature seems to be characteristic of the many-body problem. In contrast we have the results of calculations of 'gas-like' viscosity and thermal conductivity, when only two-body collisions are considered but the cross sections are calculated quantum mechanically (see, for example, Buckingham, Hamilton and Massey 1941). These give a viscosity which generally increases with rising temperature except near absolute zero, where the results depend greatly on the precise form of the interaction. The processes considered by Peierls (1929) (three-phonon Umklapp) and Landau and Khalatnikov (1949) (four-phonon) always involve the emission or absorption of small numbers of 'phonons'—processes which involve the re-arrangement of a very large number of atoms (theoretically all the atoms in the assembly are affected). Conversely, a process such as the collision or interchange of two atoms only, would be described in the Debye picture as a process involving all the normal coordinates C_s and S_s, and therefore also affecting a very large number of phonons. The relation between C_s and x_r is reminiscent of that between a variable and its Fourier transform. A process involving a single C_s involves all the x_r's and vice versa.

It therefore seems fair to regard the 'two-body' and 'Debye' contributions to the transport coefficients as fundamentally distinct from one another, and to suppose that they can, in first approximation, simply be added together. If this is so it seems that, in liquid helium II, the two types of process are of comparable importance. The 'gas-like' two-body calculations by themselves give a viscosity of the order of magnitude observed in the liquid. The 'Debye' contribution to the viscosity is very difficult to estimate numerically, and one can predict only its probable variation with temperature. To sum up, if we wish to use a model of Debye type, we have to admit the possibility of variations of the velocity of sound c_1 with density and temperature (these variations are experimentally observed). In addition we have to allow for the atomicity of the crystal lattice, which, as we have seen, implies a variation of c_1 with frequency. It is also likely that the velocity of propagation of transverse sound waves is not a constant. Thus, near the transition temperature, where the variation of c_1 with temperature begins to become rapid (Atkins and Chase 1951) a model of Landau–Debye type would contain so many adjustable constants as to be almost useless, quite apart from the fact that the large migrations of the atoms perhaps make it a poor approximation in any case.

§5. A GENERALIZATION OF THE MECHANISM OF BOSE-EINSTEIN CONDENSATION

Haselgrove and Temperley (1952) have investigated the statistical mechanics of Bose–Einstein condensation from the standpoint of the analytic theory of partition of numbers, one of their conclusions being that a sufficient condition for the occurrence of 'condensation into the lowest level' is probably that the thermodynamic probability W or $p_N(E)$, the number of ways in which the energy E can be shared among N atoms, is a maximum for *variable N and fixed E*. By Boltzmann's hypothesis, we have $S = \text{const} + k \log W$ so that the criterion for the appearance of a condensation phenomenon is

$$\left(\frac{\partial S}{\partial N}\right)_{E,V} \leqslant 0 \quad \text{or} \quad \left(\frac{\partial F}{\partial N}\right)_{T,V} \geqslant 0. \qquad \ldots\ldots(9)$$

Consider first a model of the same type as the perfect gas, in which the energy levels refer to the individual atoms, the so-called 'one-particle' type of model. At ordinary temperatures one can always gain entropy by adding another atom to the assembly at constant volume, even if no energy is added with it. This follows from the principle of equipartition of energy, valid for 'classical' assemblies. As the total energy E is reduced there will, however, come a stage when the number of ways of partitioning this energy into N non-zero parts among N atoms no longer increases with N. For N greater than this critical value, one can actually gain entropy by assigning some of the atoms no energy at all—that is to say, the most probable state will be one in which a finite fraction of the atoms are in the very lowest energy level of the assembly—the energy of which level we can always choose to be zero.

Haselgrove and Temperley (1952) have shown that this phenomenon probably occurs for a very wide class of 'one-particle' models and that it is almost certainly identical with Bose–Einstein condensation. For the perfect gas it can be formally proved that the known formulae for the energy and entropy lead to the well-known formula for the transition temperature

$$\frac{N}{V}\left(\frac{\boldsymbol{h}^2}{2\pi m \boldsymbol{k} T}\right)^{3/2} = \sum_{l=1}^{\infty} l^{-3/2} \qquad \ldots\ldots(10)$$

as a result of imposing the condition (9) $(\partial S/\partial N)_{E,V} \leqslant 0$. The formula (10) is usually derived from the consideration that the occupation of the lowest level must be finite, but condition (9) can be applied to any assembly, whereas considerations depending upon occupation numbers can usually be applied only to models of 'one-particle' type.

We therefore postulate condition (9) as the criterion for what we may call generalized Bose–Einstein condensation. Its consequence, that particles can be removed from the assembly or relegated to a state of zero energy without decreasing the total entropy, can also be exhibited in another way, which may make its physical meaning clearer. Condition (9) is equivalent to

$$g = (\partial F/\partial N)_{V,T} \geqslant 0, \qquad \ldots\ldots(11)$$

where F is the Helmholtz free energy, which states that if a particle can be reversibly removed from such an assembly under isothermal conditions and put in a state of zero free energy no mechanical work need be done. Naturally, the thermodynamic treatment gives no information on the possibility of such a transfer, it merely draws attention to the existence of two types of assembly. In a 'classical' assembly, the introduction of a further particle without energy would bring the principle of equipartition of energy into operation with a resulting increase of entropy. In a 'condensed' assembly the energy is not shared and no such entropy increase occurs. Temperley (1949) has called attention to the fact that if an adsorbing surface is in equilibrium with a condensed Bose–Einstein assembly, an adsorbed film much thicker than usual may result. The reason for this is that it is thermodynamically profitable for the film to build up as long as the energy of an adsorbed atom in the top layer is less than that of the lowest state of the Bose–Einstein assembly, whereas, in ordinary conditions, one needs an adsorption energy of several times $\boldsymbol{k}T$ to compensate for the relatively small number of 'film' states. (The same point has recently been made by Band (1951) without any acknowledgment.)

We shall now show that the Landau–Debye model of liquid helium also possesses the property (9) or (11), and, in fact, that there is a temperature below which the signs of inequality hold. This, as we shall see, results in the 'osmotic driving force' that is required to explain second sound and the thermo-mechanical effects. The perfect gas model itself is a limiting case and the signs of equality hold below the transition temperature, and the predictions of the thermo-mechanical effect and velocity of second sound follow only as limiting cases of the theory to be outlined below. In eqn. (13), for example, g is zero below the transition temperature, so that $(\partial g/\partial P)_T$ and $(\partial g/\partial T)_P$ are both zero.

For the Debye model at low temperatures, we have for the Helmholtz free energy $F = 9N\boldsymbol{k}\Theta/8 - \frac{1}{5}N\pi^4\boldsymbol{k}T(T/\Theta)^3$ where N is the number of atoms present and Θ the Debye temperature. By successive approximation we find that eqn. (11) is satisfied for temperatures below $T_c \simeq 0{\cdot}75\Theta$ if we neglect any variation of Θ with density and any change in the zero of energy such as the term V_0 in eqn. (4) and thus, at a temperature of this order, the chemical potential g changes sign, becoming positive below it. Such a change of sign has never before been thought to have any particular physical significance, in view of the fact that energies in thermodynamics are often measured from an arbitrary zero. It does, however, seem to play a part in the theory of liquid helium. In actual helium, the energy E_0 at absolute zero is probably negative (F. London 1939), the shift in the energy–zero due to the attractive forces being a little greater than the zero-point energy.

Tisza (1947) has proposed as a model of liquid helium II a Debye-like lattice in which the shear modes have undergone Bose–Einstein condensation, but he has given no actual proof of the possibility of such an effect. However, another many-body model, in which Bose–Einstein condensation would also occur, results from Bloch's 'spin-wave' treatment of ferromagnetism at low temperatures (1930). It has been remarked by many writers that this model is mathematically equivalent to the perfect gas at sufficiently low temperatures, so that it shows a transition of the same type. (This is a rigorous consequence of the assumptions on which the model is based; whether these are physically valid near the predicted transition temperature is another matter.) Thus, it is by no means a new suggestion that Bose–Einstein condensation may have analogues in the many-body type of model, and the possibility has been rigorously proved in at least one instance besides the one now being examined.

Consider a Debye lattice containing N atoms, and suppose it to be in thermodynamic equilibrium with an adsorbing surface containing M sites, each of energy ϵ. For equilibrium we may suppose that m atoms have left the lattice and are adsorbed on the sites. The entropy of m atoms on M sites is $\boldsymbol{k}\log\binom{M}{m}$ so that the free energy of the m adsorbed atoms is

$$F_{\text{ads}} = m\epsilon - \boldsymbol{k}T\log\binom{M}{m}.$$

Adding to this the free energy of $N-m$ atoms in the Debye lattice, and differentiating with respect to m, the condition for equilibrium is

$$\boldsymbol{k}T\log\left(\frac{m}{M-m}\right) + \epsilon - g = 0 \quad \text{if} \quad N \gg M. \qquad \ldots\ldots(12)$$

If, say, half the adsorbed sites are occupied, the first term in (12) will vanish, and if $(\partial F/\partial N)_{V,T}$ or g is negative, then ϵ must also be negative in order to get appreciable adsorption, whereas if it were possible to have an adsorption sites with nearly zero energy, they would begin to fill up as soon as g became positive. Expressed differently, an adsorbed film in equilibrium with a Debye lattice may be expected, below the temperature estimated above, to build up layer by layer to a thickness such that the adsorbing energy of an atom in the top layer is negligible.

This picture is in many respects similar to that of an adsorbed film in equilibrium with a condensed Bose–Einstein assembly discussed by Temperley (1949). In contrast with the perfect gas, the simple Debye model does not predict any definite transition temperature below which the film should suddenly begin to thicken, but simply suggests a slow build-up in thickness as the temperature falls. The Debye model is, however, quite capable of being modified to give a co-operative effect and make g a non-analytic function of temperature. There is clear evidence (Atkins and Chase 1951) that the approach of the transition temperature, from above or below, is heralded by a rapid fall in the measured velocity of sound c_1. If this situation is to be properly handled by a Debye-like theory, we shall have phonon energies that depend on temperature and the model must then be handled by the special technique described by Rushbrooke (1940). We do not attempt this at present, as there are few data on the form of the energy spectrum, while the particular feature of the interaction between helium atoms that is responsible for this rapid variation of c_1 with temperature has not yet been identified. A similar remark applies to the effect of the variation of c_1 or ω_0 with density. There is a corresponding variation of the zero-point energy with density, which, as many writers have pointed out, can be described as a 'zero-point pressure' which is responsible for 'blowing out' the liquid to an abnormally low density, but we are not yet in a position to predict this variation in detail from the interaction function.

§ 6. THE FOUNTAIN EFFECT

Consider now two vessels of liquid helium connected only by an adsorbed film, which we may suppose to be at a temperature that is the mean of that in the two vessels. Equation (12) shows us that the condition for the helium in either vessel to be in equilibrium with the film is $g=\epsilon_{1/2}$ where $\epsilon_{1/2}$ is the adsorption energy of a layer that is half occupied ($M=2m$). (This condition is independent of the temperature of the film itself.) Both vessels will then be in equilibrium with the film when g is equal in both vessels, which means that the pressure and temperature differences are related by

$$\frac{\Delta P}{\Delta T}=-\frac{(\partial g/\partial T)_P}{(\partial g/\partial P)_T}=\rho S, \qquad \ldots\ldots(13)$$

an equation that was originally derived by H. London (1939) by a thermodynamic cycle, assuming the existence of the film, which is capable of transferring matter without any appreciable energy transfer.

§ 7. SECOND SOUND

(i) *The Fourth Propagation Equation*

The hydrodynamics of second sound as formulated by Landau (1941) involves, besides the two ordinary equations of hydrodynamics, the assumptions that all of the entropy is carried by the normal fluid (the third propagation equation) and that the acceleration of the superfluid is proportional to the space-gradient of g (the fourth propagation equation). The third equation can be accepted as reasonable—it is in accord with the experimental fact (Kapitza 1941) that no entropy transfer occurs when helium flows through a very narrow channel. It also follows as a natural consequence of Landau's definition of ρ_n, see below (p. 503). The fourth equation is based on the thermodynamic result that, if one atom of helium is removed from a given region at constant volume and entropy, the mechanical work done is $(\partial E/\partial N)_{V,S} = (\partial F/\partial N)_{V,T} = g$ but the difficulty has been to explain why such a process can apparently be carried out reversibly only in liquid helium so that one can get an *acceleration* of superfluid proportional to gradient g. The appeal to the analogy of osmotic pressure does not really help to solve this fundamental difficulty, because, although ordinary osmotic pressure is governed by a similar thermodynamic theory, the flow which occurs in response to a difference of osmotic pressure is invariably of 'diffusion' type and is thus irreversible, the *velocity* of flow being proportional to gradient g.

The discussion we have just given seems to provide a clue to the answer. We have seen that helium atoms of very small energy, or forming part of a weakly adsorbed film, can be in thermodynamic equilibrium with a condensed Bose–Einstein assembly, or with a Debye lattice below the temperature at which $g = 0$, and the movement of such atoms from place to place could then give rise to the transfer of matter without any appreciable transfer of energy, the very process envisaged by all theories of second sound and the fountain effect. If, however, the temperature is raised so that g becomes negative such atoms of low energy can no longer be in equilibrium with the assembly, instead the assembly *shares some of its energy with them so that they become incorporated in it*, and the process then described becomes impossible. The idea of special states of low energy, 'z-states', was put forward by Mendelssohn (1945). Until now, the only known mechanism by which such low-energy states can be occupied below a certain temperature but practically empty above it is the Bose–Einstein condensation. However, the associated phenomenon of the entropy becoming a maximum as a function of N is applicable to many body models as well, and it is put forward here as the natural generalization to such models of the Bose–Einstein condensation effect. Clearly, it is not by itself a complete explanation of superfluidity—besides accounting for the *occupation* of the 'z-states' it is still necessary to show that the atoms in them can flow from place to place under a very small pressure gradient. Evidently the Rollin film, in which the top layers of atoms are weakly adsorbed and probably therefore mobile, constitutes one type of 'z-state', but the existence of second sound seems to require the existence of 'z-states' in the bulk liquid as well. I hope to make a suggestion about their nature in another paper. (The tempting hypothesis that the superfluid effects in liquid helium are all due to a 'sub-surface film' broadly similar to the Rollin film is untenable on various grounds, the most important of which are the fantastically high velocities of film flow that would be required to explain the existence of 'second sound' of quite moderate frequencies and the observed fact that the measured velocity of second sound is independent of the shape of the vessel (D. V. Osborne, private communication).) However, is quite unnecessary to postulate, for example, that at a temperature at which ρ_n/ρ is 1 %, 99 % of the atoms in the assembly are in z-states. The fallacy of supposing that ρ_n/ρ is necessarily equal to $(N - N_z)/N$ where N_z is the number of atoms in the z-states, has been pointed out by Dingle (1951). It is true for the perfect gas model, but is not true generally.

The fact that the measured chemical potential of liquid He II is decidedly negative $\simeq 10-15$ cal/mole shows that the above model is still too specialized. It is hoped to consider this point in another paper.

(ii) *Landau's Concept of Effective Mass*

Landau (1941) showed that it is possible to assign an 'effective mass' to an assembly of phonons by the following process. One imagines the assembly in thermodynamic equilibrium with a vessel the walls of which are moving with a small tangential velocity v. Relative to the walls, a difference of energy is postulated between a phonon moving in the same sense as v and an exactly similar one moving in the opposite sense, the difference in energy being assumed to be $2p_x v$, where p_x is the x-component of momentum of the phonon. Thus if the assembly of phonons is in equilibrium with the moving walls, there is a net momentum of the whole assembly. Averaging over all angles and energies, Landau shows that the net momentum is proportional to v for v small, and he thus defines an 'effective mass' of the assembly which he identifies with the mass apparently 'carried round' by a disc oscillating in liquid helium. This conception of 'effective mass' is reminiscent of the effective mass of an assembly of electrons in metals, but is definitely not equivalent to it. Landau defines effective mass as (mean momentum/velocity), whereas the effective mass of an assembly of electrons is defined, see for example Seitz (1940) as

$$\frac{\text{Total force acting on electrons}}{\text{Rate of change of electron flux}}.$$

The latter definition is inapplicable to phonons, which are unaffected by any linear external field acting on the atoms. See p. 495.

So far, Landau's theory may be regarded as an attempt to predict theoretically the apparent mass of liquid 'carried round' by an oscillating disc. His subsequent identification of the difference between this mass and the ordinary inertial mass of the liquid helium itself as the 'effective mass of superfluid' is much less easy to understand physically. Consider, for example, a toroidal vessel of liquid helium subjected successively to the processes of pushing the whole liquid forward by means of a piston, and then dragging the normal fluid back an equal distance by setting the walls in motion. Landau would interpret the final effect of these two processes as equivalent to the motion of superfluid alone, and it is easily shown that the entire entropy of an assembly of Debye waves is carried along with the walls if they are in thermal equilibrium with the assembly (Dingle 1951). Landau has therefore shown that this process of superfluid transfer would occur, if at all, without any transfer of entropy, and should thus be thermodynamically reversible, but he has not given any physical picture at all of how such flow is to take place.

(iii) *A Suggestion about the Possible Nature of Superfluid Flow*

We have suggested above that superfluid flow takes place through the medium of z-states which can, in principle, occur in any assembly. Since we do not know the nature of z-states associated with a Debye-like model, but have simply shown that if they occur, they will be occupied only below a certain temperature, we go back to the perfect gas model, the nature of whose 'z-state' is precisely known. The experimental fact that has to be explained is *not* a 'persistent velocity', analogous to the persistent current in superconductors, because no such effect has yet been observed, but the fact that superfluid flow, in films or bulk liquid, can take place, at a measurable rate, under extremely small pressure heads, of the order of a fraction of a millimetre of liquid helium or less (Daunt and Mendelssohn 1946, Bowers and Mendelssohn 1950). In this connection, it is of interest to call attention to a feature of the condensed Bose–Einstein gas that does not seem to have been explicitly noticed before, namely its extreme sensitivity to an applied external field such as gravity. It is fully realized that, in practice, this picture must be considerably modified by atomic interactions, but it seems not unreasonable to think that it has some relation to the real situation.

The energy levels of a material particle in a constant external field were found by Breit (1928). If the field is along the z-direction, we have to solve the equation

$$\frac{h^2}{8\pi^2 m}\frac{\partial^2\psi}{\partial z^2}+(E-mgz+mgz_0)\psi=0 \qquad \ldots\ldots(14)$$

the choice of the zero of z being arbitrary, and the factors of ψ involving x and y being the same as in the perfect gas. The relevant solutions of this equation are oscillatory when the kinetic energy is positive, but fall away rapidly to zero as soon as $mg(z-z_0)\geqslant E$. Insertion of numerical values shows that, for helium atoms, the characteristic distance is only of the order of 10^{-4} cm. Thus, if we choose the origin of z at the bottom of the container which we may suppose to be of dimensions of the order of 1 cm, the wave function for the lowest state will vanish at $z=0$, attain its maximum for $z\sim 10^{-4}$ cm and tend rapidly to zero for larger z, the corresponding factor of ψ being very nearly an Airy function. The next highest wave function will pass through two maxima as a function of z, and so on.

The interesting point is that, for particles of the mass of helium atoms, the lowest states are very much affected by a field of the order of the gravitational field. Without the field, the lowest state would have a factor sin $(\pi z/h)$ where h is the height of the container, whereas the presence of gravity has resulted in this state being of negligible amplitude except near the very bottom of the container. For an uncondensed gas, the probability of occupation of each state is very small, and the result of averaging over all states would lead simply to the ordinary sedimentation effect that occurs in any gas under gravity, but in a perfect gas below the Bose–Einstein condensation temperature many of the atoms enter the very lowest state. Naturally, any interactions between the atoms would modify the form of the lowest wave function very conrsideably, but it seems most unlikely that they would remove this effect altogether. In the perfect gas, if it were possible to reduce the effect of gravity by a large factor, the effect of such a reduced field on the lowest states would still be quite noticeable. From this it seems at least plausible to argue that, in actual liquid helium, the 'z-states' may be abnormally sensitive to an applied external field and one may expect a similar result for a hydrostatic pressure gradient, although it does not follow *a priori* that in a quantum assembly a pressure gradient and an applied field can simply be added together, as is possible in classical hydrodynamics.

§8. HELIUM 3

It is natural to ask whether superfluid effects are to be expected at sufficiently low temperatures. It is probable that, at sufficiently low temperatures, He_3 can be represented by a Debye model. Tomonaga (1950) has shown that this is possible for any assembly of Fermi particles whose interactions obey some not very onerous conditions, but we remain completely ignorant of the order of magnitude of the Debye temperature. (It should be possible to estimate this when data on the compressibility of He_3 become available.) In the meantime, it can certainly be said that the absence of superfluid effects in He_3 above 1° K can be understood if it should turn out that it has a Debye temperature significantly smaller than that of He_4.

We have seen that, in the region of temperatures where a Debye model is a satisfactory approximation, the type of statistics becomes unimportant. The connection between the Debye Θ and a possible transition temperature can be illustrated by considering the simplest Debye model, longitudinal sound only without dispersion. For this we should have for the transition temperature the equations

(*a*) according to Landau's criterion ρ_n/ρ

$$\rho=\frac{32\pi}{c_1{}^5\boldsymbol{h}^3}(\boldsymbol{k}T)^4\,\zeta(4) \text{ leading to } T_c\simeq 10^\circ\,\text{K};$$

(*b*) according to the criterion (11) $(\partial F/\partial N)_{T,\,V}=0$

$$T_c\simeq 0{\cdot}75\Theta\simeq 25^\circ\,\text{K},$$

if the shift in the zero of energy due to the attractive forces is ignored, see eqn. (4). The introduction of dispersion, of variation of c_1 with density and of transverse sound quanta or rotons would modify these numerical values very considerably but would not destroy the essential connection between T_c and the Debye Θ just referred to.

§ 9. THE PROBABLE FORM OF THE SPECTRUM OF HE$_4$ AT LOW ENERGIES

The transition temperature of He_4 is so much lower than these 'order of magnitude' estimates that it is quite clear that the Debye approximation is poor in the transition region. Landau (1947) assumed arbitrarily that the roton part of the spectrum could be described by $E(p)=\Delta+(p-p_0)^2/2\mu$ but Temperley (1951 a) has pointed out that such a spectrum would imply the unphysical assumption of an 'infinite' velocity of propagation of transverse sound of low frequency and also that the assumption that this velocity is zero or small compared with c_1 would be inconsistent with the observed approximate agreement of the low temperature values of c_2 and $c_1/\sqrt{3}$ (Atkins and Osborne 1950, Atkins and Chase 1951). It therefore seems advisable to assume, provisionally, that the longitudinal and transverse velocities of sound are comparable, as in an ordinary crystal lattice.

For their calculations of viscosity, Landau and Khalatnikov (1949) were mainly interested in the low energy end of the spectrum, and they supposed that Landau's detailed model which agrees reasonably with the data for both c_2 and specific heat could, for this purpose, be replaced by the simpler spectrum

$$E(p)=c_1(p-\gamma p^3). \qquad \text{......(15)}$$

They derived
$$\gamma=2{\cdot}8\times 10^{37}\,\text{g}^{-2}\,\text{cm}^{-2}\,\text{sec}^2 \qquad \text{......(16)}$$

from Landau's (1947) assumed values of Δ, μ and p_0 and they then use this value of γ to derive values of viscosity in reasonable agreement with experiment.

It is of interest to compare this value of γ with various other estimates :

(*a*) Blackman's (1935) work on the specific heat of an ideal crystal lattice suggests that significant departures from Debye's law may occur down to temperatures as low as 0·01 Θ. For a Debye temperature of 30° K (appropriate for liquid helium) the two terms in (15) would be comparable at an energy corresponding to 0·1 Θ if

$$\gamma \simeq 3{\cdot}6 \times 10^{39}\,\mathrm{g}^{-2}\,\mathrm{cm}^{-2}\,\mathrm{sec}^{2}. \qquad \ldots\ldots(17)$$

(*b*) The assumption of an energy spectrum such as (15) enables both the specific heat C_V and the velocity c_2 of second sound to be calculated in closed form. We can calculate the partition function, energy, entropy and specific heat directly, by integrating over all possible magnitudes and directions of the momentum p. For $T \ll \Theta$ the upper limit of p can be put equal to infinity. The velocity c_2 can be derived from the formula, Landau (1941),

$$c_2{}^2 = \frac{\rho_s}{\rho_n}\,\frac{S^2}{\partial S/\partial T},$$

ρ_s/ρ_n being obtained from Landau's formula (derived in the Appendix) for the effective mass of an assembly in motion

$$\frac{\rho_n}{\rho} = \frac{1}{Nm}\sum_r \frac{p_r{}^2}{\boldsymbol{k}T}\exp(\epsilon_r/\boldsymbol{k}T)/\{\exp(\epsilon_r/\boldsymbol{k}T)-1\}^2$$

where p_r is the x-component of the momentum of the rth state. We find easily

$$\frac{C_V}{V} = \frac{96\pi\boldsymbol{k}(\boldsymbol{k}T)^3}{c_1{}^3\boldsymbol{h}^3}\zeta(4)\left[1+\frac{\zeta(6)}{\zeta(4)}150\gamma\left(\frac{\boldsymbol{k}T}{c_1}\right)^2+\ldots\right] \qquad \ldots\ldots(18)$$

$$c_2{}^2 = \frac{c_1{}^2}{3}\left[1-\frac{\zeta(6)}{\zeta(4)}360\gamma\left(\frac{\boldsymbol{k}T}{c_1}\right)^2+\ldots\right] \qquad \ldots\ldots(19)$$

the second terms in the brackets representing the corrections due to dispersion of first sound, resulting from the assumed spectrum (15). Equations (18) and (19) give us two further estimates of γ as follows.

Hull *et al.* (1951) measured the specific heat down to 0·6° K and represented their data by means of the formula $C = 0{\cdot}024\,T^{6\cdot2}$ cal/g deg. Comparing this with the expected result for a Debye model with $c_1 \simeq 2{\cdot}4 \times 10^4$ cm/sec they conjectured that a change-over to the Debye law should occur at a little below 0·6° K. Kramers (1951) has recently reported measurements at still lower temperatures, confirming this conjecture. If the two terms in (18) are comparable at $T = 0{\cdot}6°$, this would mean that

$$\gamma \simeq 5 \times 10^{38}\,\mathrm{g}^{-2}\,\mathrm{cm}^{-2}\,\mathrm{sec}^{2}. \qquad \ldots\ldots(20)$$

From the data of Atkins and Osborne (1951) we see that c_2 has fallen appreciably below its limiting low-temperature value at $T \simeq 0{\cdot}4°$, and, if we choose γ of the same order as that given by (20), we see that it also makes the two terms in (19) comparable at just about this value of $T = 0{\cdot}4°$. Obviously a more detailed spectrum than (15) and the retention of more powers of T in (19) are required to describe the behaviour of c_2 at higher temperatures when the two terms in expression (19) nearly cancel.

We conclude, therefore, that a value for the constant γ certainly no larger than that suggested by Blackman's (1935) work on crystal lattices will describe both the observed low temperature behaviour of both c_2 and C_V. It would, no doubt, be possible, by introducing further terms into (15), and admitting also the existence of a similar spectrum of transverse sound quanta, to obtain complete agreement with all the data, but, as we have seen, we should have a great many adjustable constants, and a Debye model is probably a poor approximation near the transition point in any case.

This value of γ is distinctly greater than that given by Landau and Khalatnikov (1949), which, if their mechanism of viscosity is the dominant one, would mean that we cannot describe the low-temperature behaviour of all three quantities, C_V, c_2 and η in terms of a single parameter γ. However, it may well be that processes of Peierls' (1929) 'Umklapp' type (three-phonon process), would give a sufficient contribution, and the variation of η with temperature would be similar to that found by Landau and Khalatnikov (1949). The numerical constant is, however, extremely difficult to estimate and would, in any case, require knowledge of the whole frequency spectrum. It does, however, seem appropriate to consider three-phonon processes as a possible mechanism before we invoke four-phonon processes.

It is sometimes suggested that the large attentuation of first sound at low temperatures (Atkins and Chase 1951) is evidence against the validity of the phonon model near absolute zero. However, since the frequency used (of the order of 10^7 c/s) is much less than the phonon frequency corresponding to a temperature of 1° K (of the order of 10^{10} c/s) it follows that what has been demonstrated is *not* that these latter frequencies correspond to non-stationary states, but that transfer of energy *between* such states is becoming difficult. On the simple Debye model such transfers are impossible, so that once again we meet the necessity of making the model realistic by introducing a parameter such as γ, which means that the Debye modes are not quite stationary states. On this basis Khalatnikov (1950) was able to predict satisfactorily the observed trend of attenuation as a function of temperature.

We now seem to be in a position to state with fair confidence that the properties of helium II can be explained if it can be shown that the quantum-mechanical many-body problem has two consequences : (i) We have already arrived at the idea that helium II can be described by a Debye model in statistical equilibrium with some states of nearly zero energy. Such a picture is fully in keeping with the abnormal weakness of the atomic interactions and the large lattice spacing. (ii) We must still ask whether this picture leads to superfluidity and to the apparent presence of two distinct densities.

It is clear that, if a Debye model is a valid approximation at all, we must admit to the existence within a given volume of two quite distinct momenta, even in an ordinary liquid.

1. The mean momentum P_1 or $\rho_1 v_1$ associated with the Debye waves. This momentum must be assigned physical reality, because, by absorption of the phonons, it can be transferred to material particles, as is pointed out in the Appendix. It is there shown further that, by considering the Debye waves to be in statistical equilibrium with a moving body, we can arrive at separate definitions of ρ_1 and v_1, the effective density and stream velocity of the Debye waves, equivalent to Landau's definitions (1941).
2. The total momentum of the centre of gravity of the Debye lattice, described for instance by the coordinate C_0 in eqn. (4), together with the momentum, if any, of the atoms in states of nearly zero energy. This total momentum we call ρV, where ρ is the *total* density of the assembly.

Comparison with the two-fluid picture shows that ρ_1 must be identified with the density ρ_n of the normal fluid in order to obtain the correct variation of c_2 with temperature. Further, we must identify v_1 with v_n, because of the experimental fact that the entropy is carried by the normal fluid and Dingle's result (1951) that the entropy of a Debye assembly is transported with the velocity v_1. Since sound waves never transport any mass, we must identify the rate of transport of mass, $\rho_n v_n + \rho_s v_s$ with ρV so that we have $v_s = (\rho V - \rho_n v_n)/(\rho - \rho_n)$. On this basis, we can derive equations in first approximation equivalent to those of the two-fluid theory, using the postulates of conservation of mass, of rate of change of material momentum being proportional to gradient of hydrostatic pressure, of conservation of entropy (transported with velocity v_n) and finally of rate of change of momentum of Debye waves being proportional to gradient of sound pressure. Sound pressure is defined in a manner analogous to radiation pressure (Ward and Wilks 1951).

Superfluid effects manifest themselves, not through v_s being different from zero but through v_s being different from v_n, that is, through v_1 being different from V. Now on the simple Debye model, there is no coupling between any of the C_s and C_0, but we have seen that we have to admit such coupling between the C_s's in an actual liquid in order to explain the finite viscosity and thermal conductivity. Furthermore the absence of superfluid effects in ordinary liquids indicates a reasonably strong coupling between C_0 and the remaining C_s's. In the Appendix, it is shown that Debye waves will be in statistical equilibrium with a body only if its velocity of translation is equal to v_1, and we may apply this result, in particular, to the motion of the centre of gravity of the lattice. We must suppose that in an ordinary liquid the coupling (due to the cubic and higher terms in the interaction energy) is strong enough to maintain statistical equilibrium between C_0 and the other C_s's. In liquid helium II we can explain a difference between v_1 and V in one of two ways: (*a*) the presence of atoms in *z*-states means that V may no longer be quite equal to the velocity of the centre of gravity of the lattice, which velocity is still constrained by the coupling to be equal to v_1. (*b*) The coupling of C_0 to the other C_s's suddenly becomes too weak to maintain statistical equilibrium.

On present knowledge we cannot exclude either of these possibilities, but (*a*) seems far more plausible than (*b*) in view of the sudden onset of superfluidity and the known existence of a thick film.

CONCLUSIONS

We conclude that the Debye model is a satisfactory approximation to He_4 at sufficiently low temperatures, and that its range of application can probably be extended somewhat by taking account of the probable presence of transverse waves, and the variations of the effective propagation velocities with density, temperature and frequency. It is not yet possible to say whether it will ultimately prove to be a useful approximation in the region of the transition temperature.

The Bose–Einstein gas-like model has been very valuable in gaining physical insight into the behaviour of liquid helium II and it seems very likely that two of its features, the condensation phenomenon, and the sensitivity of the lowest states to an applied field of force have analogues in the many-body model, even when the interactions are very strong, and that both these analogues will find a place in the theory of liquid helium II.

Many of the properties of helium II follow from a model in which a Debye lattice is in statistical equilibrium with atoms in states of small energy and entropy. This model would have a transition temperature above which the latter states would be unoccupied.

REFERENCES

Andronikashvili, E. L., 1948, *J. Exp. Theor. Phys., U.S.S.R.*, **18,** 424 and 429.

Atkins, K. R., 1951, *Proc. Phys. Soc.* A, **64,** 833.

Atkins, K. R., and Chase, C. E., 1951, *Proc. Phys. Soc.* A, **64,** 826.

Atkins, K. R., and Osborne, D. V., 1950, *Phil. Mag.*, **41,** 1078.

Auluck, F. C., and Kothari, D. S., 1945, *Proc. Camb. Phil. Soc.*, **41,** 175.

Band, W., 1951, *J. Chem. Phys.*, **19,** 435.

Blackman, M., 1935, *Proc. Roy. Soc.* A, **149,** 117.

Bloch, F., 1930, *Z. Phys.*, **61,** 206.

Bowers, R., and Mendelssohn, K., 1950, *Proc. Phys. Soc.*, **63,** 178.

Breit, G., 1928, *Phys. Rev.*, **32,** 273.

Brillouin, L., 1922, *J. Phys. Radium*, **6** (T.3), 362.

Buckingham, R. A., Hamilton, J., and Massey, H. S. W., 1941, *Proc. Roy. Soc.* A, **179,** 103.

Daunt, J. G., and Mendelssohn, K., 1946, *Nature, Lond.*, **157,** 839.

Dingle, R. B., 1951, *Phil. Mag.*, **42,** 1080.

Gorter, C. J., and Mellink, J. H., 1949, *Physica*, **15,** 285.

Gorter, C. J., Mellink, J. H., and Castelejn, P. W., 1950, *Physica*, **16,** 113.

Haselgrove, C. B., and Temperley, H. N. V., 1952, in the course of publication.

Hull, R. A., Wilkinson, K. R., and Wilks, J., 1951, *Proc. Phys. Soc.* A, **64,** 379.

Khalatnikov, I. M., 1950, *J. Exp. Theor. Phys., U.S.S.R.*, **20,** 243.

Kapitza, P. L., 1941, *J. Phys., U.S.S.R.*, **4,** 181.

Kramers, H. C., 1951, *Oxford Conference on Low Temperatures* (Clarendon Laboratory, Oxford).

Landau, L. D., 1941, *J. Phys., U.S.S.R.*, **5,** 71 ; 1947, *Ibid.*, **11,** 91.

Landau, L. D., and Khalatnikov, I. M., 1949, *J. Exp. Theor. Phys., U.S.S.R.*, **19,** 637 and 709.

London, F., 1939, *J. Phys. Chem.*, **43,** 49.

London, H., 1939, *Proc. Roy. Soc.* A, **171,** 484.

Mendelssohn, K., 1945, *Proc. Phys. Soc.*, **57,** 371.

Peierls, R., 1929, *Ann. Phys., Lpz.*, (5), **3,** 1055.

Rushbrooke, G. S., 1940, *Trans. Faraday Soc.*, **36,** 1035; 1949, *Introduction to Statistical Mechanics* (Oxford : University Press), Chap. 3, para. 1.

Seitz, F., 1940, *Modern Theory of Solids* (London : McGraw-Hill), p. 316.

Simon, F. E., 1934, *Nature, Lond.*, **133,** 529.

Temperley, H. N. V., 1947, *Oxford Conference on Low Temperatures*; 1949, *Proc. Roy. Soc.* A, **198,** 438; 1951 a, *Phil. Mag.*, **42,** 74; 1951 b, *Brussels Conference on Ultrasonics.*

Tisza, L., 1947, *Phys. Rev.*, **72,** 838.

Tomonaga, S., 1950, *Prog. Theor. Phys.*, **5,** 544.

Wilson, A. H., 1936, *The Theory of Metals* (Cambridge : University Press).

Ward, J., and Wilks, J., 1951, *Phil. Mag.*, **42,** 314.

Wergeland, H., 1947, *K. Norsk. Vidensk. Selsk. Förh.*, **19,** No. 22.

APPENDIX

Revised Calculation of the Effective Mass of a Moving Assembly

We re-apply the methods of statistical mechanics to an assembly in which we specify linear momentum as well as energy. (The extension to angular momentum is immediate.) We suppose that each level in the assembly may be occupied any number of times, and that to the rth level we may assign an energy ϵ_r and an x-momentum p_r, the latter quantity being the change in momentum produced at a wall if the occupation number of the rth level changes by unity as the result of a collision. (Since the collision of excitations such as those described by eqns. (4) and (5) with a *wall* does not yet seem to have been considered explicitly we can, strictly speaking, only infer the relation $\epsilon = c|p|$ for phonons from the correspondence principle. We shall however assume it generally. With this definition of the momentum of phonons, momentum is conserved when *electrons* collide with a crystal lattice.*

The number of ways in which one can realize a state of total energy E and total x-momentum P is the coefficient of $y^P z^E$ in the generating function $\Pi_r(1-y^{p_r}z^{\epsilon_r})^{-1}$. If the total number of excitations were fixed (e.g. if the levels referred to individual particles) we could introduce a third selector variable. Then, applying the saddle-point method, we have for the entropy S

$$S \sim \boldsymbol{k}\Sigma_r \log(1-\phi^{p_r}\theta^{\epsilon_r})^{-1} - \boldsymbol{k}P\log\phi - \boldsymbol{k}E\log\theta, \qquad \ldots\ldots(\mathrm{A}\,1)$$

where $y=\phi$, $z=\theta$ is the position of the saddle-point. By general theory we may infer that $\theta = \exp(-1/\boldsymbol{k}T)$. For the free energy we have

$$F = \boldsymbol{k}T\Sigma_r \log\{1-\phi^{p_r}\exp(-\epsilon_r/\boldsymbol{k}T)\} + \boldsymbol{k}TP\log\phi, \qquad \ldots\ldots(\mathrm{A}\,2)$$

where ϕ is determined by

$$P_1 = \sum_r \frac{p_r\phi^{p_r}\exp(-\epsilon_r/\boldsymbol{k}T)}{1-\phi^{p_r}\exp(-\epsilon_r/\boldsymbol{k}T)}. \qquad \ldots\ldots(\mathrm{A}\,3)$$

We now assume that, to every level with a given p_r corresponds a level with the same energy but an x-momentum $-p_r$. This follows quite generally from symmetry considerations. Now put $\phi = e^{\beta}$. We then have

$$P_1 = \sum_r \frac{p_r\exp(\epsilon_r/\boldsymbol{k}T)\sinh(\beta p_r)}{1+\exp(2\epsilon_r/\boldsymbol{k}T) - 2\exp(\epsilon_r/\boldsymbol{k}T)\cosh(\beta p_r)}, \qquad \ldots\ldots(\mathrm{A}\,4)$$

$$F = \frac{\boldsymbol{k}T}{2}\sum_r \log\{1+\exp(2\epsilon_r/\boldsymbol{k}T) - 2\exp(\epsilon_r/\boldsymbol{k}T)\cosh(\beta p_r)\} + \boldsymbol{k}TP_1\beta.$$

For small P, β is also small, and these expressions reduce to

$$P = \beta\sum_r \frac{p_r^2\exp(\epsilon_r/\boldsymbol{k}T)}{\{\exp(\epsilon_r/\boldsymbol{k}T)-1\}^2} + \mathrm{O}(\beta^3),$$

$$F = \boldsymbol{k}T\sum_r \log\{1-\exp(-\epsilon_r/\boldsymbol{k}T)\} + \frac{\boldsymbol{k}T}{2}\beta^2\sum_r \frac{\exp(\epsilon_r/\boldsymbol{k}T)}{\{\exp(\epsilon_r/\boldsymbol{k}T)-1\}^2} + \mathrm{O}(\beta^4).$$

We now determine the value of β so that this assembly shall be in statistical equilibrium with a body of mass M, moving with a velocity v_1 in the x-direction. The total momentum of assembly and body is $P+Mv$ which is constant, while the total free energy is

$$\boldsymbol{k}T\sum_r \log\{1-\exp(-\epsilon_r/\boldsymbol{k}T)\} + \frac{\boldsymbol{k}T\beta^2}{2}\sum_r \frac{\exp(\epsilon_r/\boldsymbol{k}T)}{\{\exp(\epsilon_r/\boldsymbol{k}T)-1\}^2} + \tfrac{1}{2}Mv_1^2.$$

β has to be chosen so that F is a minimum, subject to the momentum being conserved, which requires that $\beta = v_1/\boldsymbol{k}T$. The mean momentum of the assembly is then

$$\frac{v_1}{\boldsymbol{k}T}\sum_r \frac{p_r^2\exp(\epsilon_r/\boldsymbol{k}T)}{\{\exp(\epsilon_r/\boldsymbol{k}T)-1\}^2}$$

which gives an 'effective mass'

$$\frac{1}{\boldsymbol{k}T}\sum_r \frac{p_r^2\exp(\epsilon_r/\boldsymbol{k}T)}{\{\exp(\epsilon_r/\boldsymbol{k}T)-1\}^2}$$

---

* See, for example, Wilson (1936), chap. 6.

which is equivalent to Landau's expression $-\Sigma_r p_r^2(\partial/\partial\epsilon_r)\{\exp(\epsilon_r/\boldsymbol{k}T)-1\}^{-1}$ and we can complete the calculation for the phonon model by putting $\epsilon=c|p|$, $p_r=|p|\cos\theta$ and integrating over all angles and all positive values of $|p|$.

This treatment still assumes that a phonon has momentum that is transferable to material particles, but avoids Landau's dubious assumption that the energy of such a phonon is different for different observers. The treatment on p. 495 indeed seems to show that this assumption is incorrect.

It is of interest to recall an old result of Brillouin's (1922) who concluded that, if an assembly consisting of Debye waves were sheared with uniform velocity, the sound waves would be refracted in such a way as to tend to *increase* the momentum content of the faster-moving regions of the assembly. This shows that a physical mechanism, by which a moving assembly can approach the state of statistical equilibrium calculated in (A4), actually exists. We also notice that β, and therefore the mean momentum P, both depend uniquely on v_1. This means that an assembly of Debye waves cannot be in statistical equilibrium with *two* bodies, e.g. a wall and the centre of gravity of the crystal lattice, unless they are both moving with the same velocity.

.A
5
2

The Thermal Conductivity of Liquid Helium I

BY R. BOWERS*
The Clarendon Laboratory, Oxford

Communicated by K. Mendelssohn; MS. received 14*th September* 1951

ABSTRACT. The thermal conductivity of liquid helium has been measured between 2·2 and 4° K. It is found to increase with temperature over the whole range and shows no sign of anomalous behaviour just above the λ-point.

This variation is compared with that of the viscosity and entropy in the same temperature range.

§ 1. INTRODUCTION

IN an earlier paper (Bowers and Mendelssohn 1950) we discussed the fact that the fall in entropy associated with the lambda transition begins at a slightly higher temperature than the actual transition point of 2·19° K. It was thought that this preliminary entropy drop might manifest itself in the transport properties of liquid helium I within about ½ degree of the λ-point. In the earlier work, a fall in the viscosity in this temperature region was indeed observed. The present paper describes the measurement of the thermal conductivity of liquid helium I.†

As was the case with the viscosity determinations, earlier work below 2·19° K (e.g. Keesom, Saris and Mayer 1940, Allen and Ganz 1939) gave no indication that high heat flows would persist above the λ-point. However, the type of heat flow observed by these authors in helium II was of a very anomalous kind which cannot be characterized by a thermal conductivity in the normal sense.

---

[*] Now at the Institute for the Study of Metals, Chicago.

[†] A short preliminary note of the results of this work has been published earlier (Bowers and Mendelssohn 1951). More recently, a determination of the heat conductivity of He I has been reported by Grenier (1951), whose results are in satisfactory agreement with ours.

Our work on the viscosity suggested that even though the anomalous type of transport vanished at the λ-point, there still remains the possibility that the normal heat conductivity could change markedly within ½ degree of the λ-point.

The only previous determination of the heat conductivity of liquid helium I was that of Keesom and Keesom (1936). They measured a single point at 3·3° K and obtained the value 6×10^{-5} cal/deg. cm sec, which is of the order of the thermal conductivity of a gas at room temperature. However, Pellam and Squire (1947) have suggested that there might be a considerable variation of the thermal conductivity of helium I with temperature, the conductivity increasing with decreasing temperature. This suggestion arose from their study of ultrasonic attenuation in liquid helium.

§2. METHOD

The measurement was carried out on a vertical cylinder of the liquid. Heat was supplied to the top of the cylinder and was conducted down to a heat sink at the bottom of the cylinder which could be maintained at a constant temperature. Two thermometers were fixed at different heights within the cylinder, mounted parallel to its base. For zero side loss of heat from the cylinder, the thermal conductivity K may be calculated from $Q=KA(\theta_1-\theta_2)/L$ where $Q=i^2R=$the rate of production of heat in the heating plate, θ_1 and θ_2 are the equilibrium readings of the upper and lower thermometers for the heat flow Q, A is the area of cross section of the cylinder, and L is the distance between the thermometers.

The actual form of the conductivity cell is shown in fig. 1 (*a*). A is a copper base on which were mounted the two resistance thermometers C and D. The heater B and the resistance thermometers were mounted parallel to the top of the copper base, being supported by three tubes of german silver of ½ mm diameter and $\frac{1}{10}$ mm wall thickness. The heater was a coil of Eureka wire of about 110 ohms and the heat generated in it was distributed uniformly over the circular cross section by the plate of copper to which it was attached. The thermometers, which were required to measure the temperature in a plane of circular cross section, consisted of leaded-brass wire wound over a former of amber. About 20 cm of resistance wire was required in order to obtain the necessary sensitivity and this wire was wound on to the former in the manner shown in fig. 1 (*b*). It was unavoidable that the wire, which ideally should have been entirely accommodated in one plane, tended to bow on the former. However, since the bowing was symmetrical about the plane of the former, no appreciable error was introduced and the recorded temperature could be taken as equal to that in the annular plane. The leads for the thermometers and for the heater (three leads for each thermometer and two for the heater) passed out of the cell through tube E and then through eight 'Covar' glass-to-metal seals soldered into the copper base. The heater and thermometers were enclosed by an inverted glass Dewar vessel G which ensured that all the heat generated in the heater flowed down to the heat sink, i.e. it prevented side loss.

The conductivity cell was suspended in the experimental space of a small scale Linde liquefier (Daunt and Mendelssohn 1948). During an experiment, the apparatus was surrounded by liquid helium, and liquid helium could be condensed into the measuring cell through tubes E and H.

Measurements of the thermal conductivity of liquid helium I present a number of peculiar difficulties which result from a combination of high specific heat, very low heat conductivity and an extremely high fluid mobility. Since a detailed discussion of these difficulties has been presented elsewhere (Bowers 1951), the experimental problems involved need only be mentioned briefly in this paper. It was found that even small vibrations in the laboratory were sufficient to produce convection currents inside the cell which made accurate determinations of equilibrium temperatures difficult. The experiments had therefore to be carried out at night, but even under these conditions, vibration represented a serious cause of random experimental error. A further difficulty was introduced by the fact that the heat conductivity of any suitable container material, even glass, is very much larger than that of liquid helium I (ratio of 3·5 to 1 in the case of ordinary soft glass at 3° K). It is therefore essential to use a cylinder of helium of the maximum radius possible and the thinnest container wall in order to prevent an excessive quantity of heat flowing down the container. A limitation on the size of apparatus which can be placed in our liquefier-cryostat led to the fact that about 50% of the heat from the heating plate flowed down the inner glass wall of the container Dewar. Thus a correction of this magnitude had to be applied to the measured conductivity of the cell in order to obtain the conductivity of the liquid helium itself.

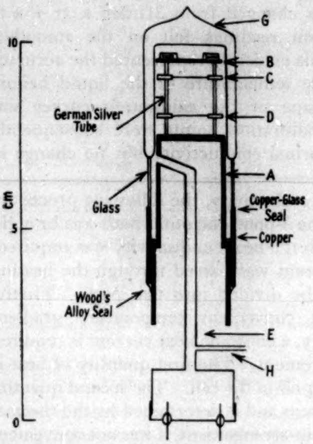


Fig. 1 (*a*). The conductivity cell.

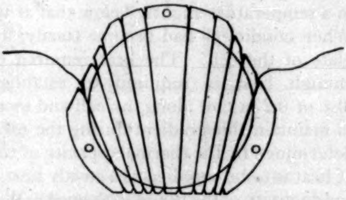


Fig. 1 (*b*). Construction of thermometer.

The experimental procedure adopted for the measurements was as follows. As helium was being liquefied into the cryostat containing the conductivity cell, a pressure of about 100 cm Hg was maintained in the liquid in the conductivity cell. During the filling of the cryostat, the cell was found to fill to about two-thirds of its total capacity with liquid. The cell was completely filled by pumping the outer bath below the λ-point, when the condensation became very rapid and all temperature gradients in the liquid disappeared. The liquid in the cell was then brought to the temperature required for a determination of the conductivity or for a calibration of the thermometers by warming the outer bath to this temperature. The temperature of the liquid in the cell followed that of the outer bath quite rapidly because the principal process of heat flow involved was that of convection. Due to a combination of low viscosity and large expansion coefficient in liquid helium I, thermal convection currents can equalize temperatures rapidly.

As the liquid in the cell warmed up from below the λ-point, the pressure exerted on it was maintained at a steady value of 90 cm Hg. This pressure, which always exceeded the vapour pressure, was maintained throughout the experiment so as to prevent any evaporation of liquid inside the cell. Moreover, during a determination of the conductivity, the cell was regularly inspected for the appearance of bubbles.

The resistance of the thermometers was calibrated for a number of different temperatures determined from the vapour pressure of the liquid in the outer bath. The sensitivity of the thermometers changed from 3Ω/deg. K at 4° K to $\frac{1}{3}\Omega$/deg. K at 2·2° K. Individual calibration readings fell on the smoothed calibration curve to within about $\frac{1}{100}$° K. This evidently represented the accuracy with which we can determine the absolute temperature of the liquid helium within the cell. Although the main shape of the calibration curves was determined in these initial experiments, calibration points were subsequently taken before each determination of the thermal conductivity but no change in the calibrations was found.

For making a measurement of the heat conductivity, the following procedure was adopted. Having filled the cell below the λ-point the outer bath was brought to a temperature slightly below that at which the heat conductivity was required. When conditions had become steady, a current was passed through the heating plate of the cell. The heat required can be divided into two parts. Firstly, enough heat is required to establish a convenient temperature gradient (0·1 or 0·2° K/cm) along the cell and secondly, a constant heat current is required to maintain this gradient during the measurement. The first quantity of heat is determined by the thermal capacity of the liquid in the cell. The second quantity of heat is to be supplied as a steady heat current and is determined by the thermal conductivity of the liquid in the cell. With our arrangement, it was not convenient to supply the heat necessary for establishing the temperature gradient using the same steady current required to maintain it. The thermal capacity of the liquid is so large and its heat conductivity so small that the steady flow heat current would have taken four or five hours to supply the total heat necessary for equilibrium. It was clearly more convenient to put in the heat required for the establishment of the temperature gradient in a short time, using a large current through the heater for a few minutes and then reducing the current to the value required to maintain the gradient of temperature. Using this technique, equilibrium could be reached in about 2 hours and readings were continued for a further two hours in order to establish accurately the temperature difference between the thermometers.

§ 3. RESULTS

A series of observations was made at temperatures between 2·2° K and 4° K. The results of these measurements are shown in fig. 2. In this figure we have plotted an effective conductivity K_e of the cell filled with helium between the two thermometers. The values of K_e together with the temperature differences used are given in the table. K_e is not the conductivity of the helium because it contains a large contribution due to conduction down the inner wall of the Dewar and a small contribution due to conduction down the german silver supports and the Eureka wires. The correction necessary to allow for this residual conductivity is also shown in fig. 2. This correction was calculated from measurements of the conductivity of a specimen of glass of the same batch as that of the Dewar vessel; it has been found that the conductivity does not vary

Details of Heat Conductivity Measurements

	ΔT	T	K_e	K
(mA)	(° K)	(° K)	($\times 10^{-5}$ cal/deg. cm sec)	
0·50	0·047	2·780	8·20	4·25
0·70	0·104	2·824	7·44	3·42
0·75	0·111	2·719	7·98	4·13
1·01	0·183	2·786	8·79	4·82
0·90	0·166	2·573	7·69	4·09
0·80	0·171	2·425	5·91	2·54
1·15	0·216	3·118	9·65	5·15
0·65	0·105	2·292	6·38	3·23
0·88	0·161	2·353	7·63	4·38
0·91	0·202	2·505	6·45	3·00
0·75	0·120	2·320	7·40	4·20
0·86	0·177	2·352	6·59	3·34
0·70	0·131	2·450	5·90	2·50
0·70	0·121	2·281	6·39	3·26
0·55	0·076	2·251	6·05	3·00
1·25	0·221	3·655	11·0	5·65
1·15	0·165	3·941	12·7	6·90
1·05	0·167	3·097	10·4	5·95
0·75	0·121	2·450	7·33	3·91
0·97	0·189	2·515	8·30	4·80
0·87	0·184	2·444	6·52	3·12
1·12	0·216	3·380	9·15	4·22
1·24	0·196	3·973	12·4	6·57

i = steady current through heater (resistance = 109Ω), ΔT = resulting temperature difference, T = mean temperature of determination.

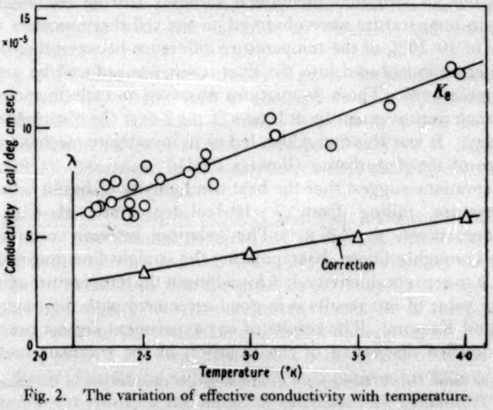


Fig. 2. The variation of effective conductivity with temperature.

appreciably from one sample to another.* The evaluation of the residual heat flow by a direct method involving evacuation of the measuring cell was not possible because the heater and thermometers were not connected directly to the glass. The use of a gas at low pressures was also not practicable because of the long equilibrium times involved in this measurement. However, it should be remembered that our main interest in this work was the variation of the conductivity of helium I with temperature, particularly just above the λ-point, and not its exact absolute magnitude. Thus a precise evaluation of this correction is not of the greatest importance.

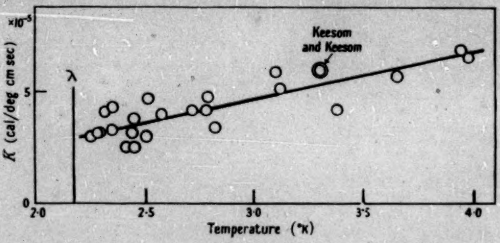


Fig. 3. The variation of heta conductivity of helium I with temperature.

The final curve of the heat conductivity for liquid helium I is shown in fig. 3.

§4. DISCUSSION OF RESULTS

The scatter of points in fig. 2 is rather large; the mean scatter of points from the straight line drawn is 6%. There does not appear to be any correlation between the deviation of any point from the line and the temperature gradient used in its measurement. We have no evidence that would suggest that the heat flow was non-classical, so that we regard the scatter of points as purely experimental error. Probably the most important single contribution to this was the disturbance due to vibration mentioned earlier. During the determinations, fluctuations in temperature were observed on the cell thermometers which were of the order of 10–20% of the temperature difference between them. However, the random error introduced into the final result was reduced by averaging the observed fluctuations. These fluctuations observed in experiments carried out at night were an improvement by at least a factor 2 over the disturbance observed during the day. It was this fact which led us to investigate mechanical vibration as the source of the disturbance (Bowers 1951).

Our observations suggest that the heat conductivity of liquid helium I varies with temperature, falling from 7×10^{-5} cal/deg. cm sec at 4° K to about 3×10^{-5} cal/deg. cm sec at 2·2° K. The relation between conductivity and temperature is roughly linear. Extrapolating the straight line drawn through the points of fig. 3 to zero conductivity, it is found to cut the temperature axis at 0·75° K. The absolute value of our results is in good agreement with the single value due to Keesom and Keesom. The results of our experiment are not precise enough to allow a detailed discussion of the variation of the thermal conductivity of liquid helium I with temperature. It should be recalled, however, that we were primarily interested in the question of whether the conductivity changes in a way which would suggest a preparation for the superfluid state as the λ-point was approached. Our results do not show any marked anomaly near the λ-point, although the scatter is too large to rule out changes in the conductivity of the order of 10%. In no case did we observe very high values of the conductivity just above the λ-point, so that the effective conductivity of the liquid helium must change very sharply on passing through the λ-point.

---

* We wish to thank Dr. R. Berman for determining the conductivity of the glass required for evaluating this correction.

In order to get some further qualitative information on this sudden change at the λ-point, a short experiment was carried out which involved heating the cell through the λ-point. During this experiment the temperature difference between the two thermometers was measured as the liquid helium was slowly heated from below the λ-point. It was observed that a temperature difference between the thermometers began to appear within a few thousandths of a degree above the λ-point. This is further evidence for our view that the change in effective conductivity is very rapid at this point.

§ 5. CONCLUSION

In order to analyse the results of this experiment we shall consider to what degree they are consistent with the earlier observations on the viscosity of liquid helium I and with the known shape of the entropy curve (Keesom and Keesom 1935). The viscosity showed a preliminary fall just above the λ-point in the same temperature region in which the entropy showed its first anomalous decrease. So that both the viscosity and the entropy just above the λ-point indicate some preparation for the superfluid state. Our earlier work (Bowers and Mendelssohn 1949, 1950) showed that the observed fall in the viscosity really represented a fall in the true viscosity of helium I, there being no sign of a flow with the anomalous character as is observed below the λ-point. Thus the preparation for the superfluid state took the form of a change in the well-defined viscosity and could not be explained by the inception of a small degree of superfluidity. The heat conductivity, however, does not appear to show a similar preparation for the superfluid state, but such an observation is not necessarily inconsistent with the variation of viscosity and entropy in this temperature region.

According to the classical kinetic theory of gases, the thermal conductivity K, the viscosity η and the specific heat at constant volume C_v are related by the relation $K = \text{const.}\ \eta C_v$. Although helium I is a liquid, it does have a number of gas-like properties. Due to its large zero-point energy, its density is quite low (Simon 1934). Moreover, its viscosity increases as the temperature increases, a property characteristic of gases.

The dependence of K on the product ηC_v may provide an explanation for the insensitivity of the thermal conductivity to the approach of the superfluid state. As the temperature is reduced below 2·6° K, the viscosity decreases while the specific heat increases, so that the product ηC_v may not change appreciably in this temperature region. A more quantitative analysis of the product ηC_v is hardly justified, considering the crude gas model underlying it. It is, however, worth mentioning that if the mean free path λ is calculated from the gas relation $K = \frac{1}{3}\rho\bar{c}\lambda C_v$ then a value of the order of 10^{-7} cm is obtained. This is of the order of the interatomic distance in the liquid as calculated by London (1936).

ACKNOWLEDGMENTS

The author wishes to thank Dr. K. Mendelssohn for the interest he has taken in this work and Mr. D. F. Brewer for his assistance with the experiments. This research was carried out while the author was receiving a grant from the Department of Scientific and Industrial Research, for which grateful acknowledgment is made.

REFERENCES

ALLEN, J. F., and GANZ, E., 1939, *Proc. Roy. Soc.* A, **171,** 242.
BOWERS, R., 1951, *Thesis*, University of Oxford.
BOWERS, R., and MENDELSSOHN, K., 1949, *Proc. Phys. Soc.* A, **62,** 394; 1950, *Proc. Roy. Soc.* A, **204,** 366; 1951, *Nature, Lond.*, **167,** 111.
DAUNT, J. G., and MENDELSSOHN, K., 1948, *J. Sci. Instrum.*, **25,** 318.
GRENIER, C., 1951, *Phys. Rev.*, **83,** 598.
KEESOM, W. H., and KEESOM, A. P., 1935, *Physica, 's Grav.*, **2,** 557; 1936. *Ibid.*, **3,** 359.
KEESOM, W. H., SARIS, B. F., and MEYER, L., 1940, *Physica, 's Grav.*, **7,** 817.
LONDON, F., 1936, *Proc. Roy. Soc.* A, **153,** 576.
PELLAM, J. R., and SQUIRE, C. F., 1947, *Phys. Rev.*, **72,** 1245.
SIMON, F. E., 1934, *Nature, Lond.*, **133,** 529.

Heat Transport in Lead–Bismuth Alloys

BY J. L. OLSEN
Clarendon Laboratory, Oxford

Communicated by F. E. Simon; *MS. received 3rd March* 1952

ABSTRACT. The variation with temperature and with magnetic field of the heat conductivity of a series of lead–bismuth alloys has been measured. As the concentration of bismuth is increased the heat conductivity in the normal state falls below that in the superconducting state, contrary to the case in pure superconductors. Estimates have been made from the experimental data of the processes contributing to the heat conductivity, and of the resistances due to the various scattering mechanisms limiting these conductivities.

§1. INTRODUCTION

PRELIMINARY results of a series of measurements of the thermal conductivities of superconductors have already been published (Mendelssohn and Olsen 1950 a, b, c). These results showed that processes other than the conduction by the electrons must be important components of the total thermal conductivity in the superconducting state. It is the object of this paper to present fuller experimental data and to attempt a quantitative analysis of the observed effects.

§2. APPARATUS

The measuring part of the apparatus is shown in fig. 1. The specimen S was placed horizontally and connected thermally to the expansion chamber C of an expansion liquefier (Simon 1932) by the copper connecting rod K. The temperature gradient produced by heating electrically with the heater W was measured by two gas thermometers T′ and T″. The pressures in these thermometers were transmitted to an external manometer system by the capillaries M. The specimen and thermometers were surrounded by the radiation shield R, whose interior communicated with the vacuum space of the liquefier.

The temperature of the expansion chamber could be kept at any temperature between 20° K and about 10° K by allowing hydrogen in a container (not shown in the diagram) attached to the expansion chamber to evaporate at an appropriate pressure. Temperatures between this and the critical point of helium could be obtained by allowing the helium in the expansion chamber to expand at a suitable rate (MacDonald and Mendelssohn 1950). Lower temperatures were obtained by letting the helium boil under a reduced pressure.

The thermometers were filled to a pressure of between 15 and 75 cm Hg when the apparatus was at 20° K, and subsequent pressure differences between the two thermometers were measured by a differential oil manometer. Since the difference in height of the two oil levels could be read by eye to about 0·2 mm, changes in temperature differences of less than 0·001° could be read with the higher filling pressures in the thermometers. The temperature differences set up by the heater were usually made of the order of 0·1°, and there was consequently a possibility of obtaining the thermal conductivities with an accuracy within 1%.

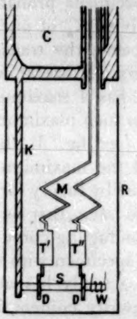


Fig. 1. Arrangement of specimen and thermometers.

The conduction along the thermometer capillaries and supports was too small to be measured in the absence of a specimen, and has therefore been calculated. It is found to lie below 1% of the conduction of the specimen in all cases. Departure of the helium in the gas thermometers from the ideal gas laws causes a negligible error in the temperature difference, but a correction has been made to the absolute temperatures for this effect. The dead volume of the capillaries introduces an error which is always difficult to correct for, but in our case this is less than 1%. One of the most dangerous errors in thermal conductivity measurements, because it may pass unnoticed, is that due to conduction through the residual gas in the vacuum surrounding the specimen. The use of a combined liquefier–cryostat has the advantage that this danger is completely eliminated, since a deterioration of the vacuum is immediately shown by an abnormal evaporation of helium from the cryostat. This evaporation would become obvious when the conductivity of the residual gas reached about 10^{-6} watt/deg cm which would correspond to an error of 5×10^{-4} watt/deg cm in the measured conductivity of a specimen of usual size.

The specimens were made from pure lead obtained from Johnson, Matthey (Lab. No. 1932) to some of which 0·02, 0·1, 0·2, 0·5, 10 and 30% of bismuth (Lab. No. 4213) from the same source had been added. The shape of the specimens is seen in fig. 1. The two copper discs D, to which the thermometers were eventually attached with solder, were a part of the mould. They were wiped inside with lead to make them adhere to the specimen as it solidified. This avoided the falsification of magnetic field values by persistent currents set up in rings of solder.

§ 3. VARIATION WITH TEMPERATURE

(i) *Experimental*

In fig. 2 the thermal conductivities of two specimens of pure lead are shown. These specimens were cast in vacuum direct from the ingots obtained from the makers. They may be expected to be polycrystalline and less pure than the triply recrystallized specimens of de Haas and Rademakers (1940, Rademakers 1949). This is also in agreement with the fact that we find a maximum in the conductivity which is three times lower than theirs. While their specimens were stated to be single crystals, both our specimens probably had a fairly small grain size.* The superconducting conductivity K_s of our specimens departs sharply from the normal conductivity K_n below the transition temperature T_c just as has been found by previous workers. However, while they found that it falls to a minimum and then rises and has a maximum at about 3·5° K, below which temperature K_s falls steeply, no such maximum was observed in our specimens.

When impurity is added (see fig. 3) the absolute value of the normal conductivity is decreased, and the maximum in the conductivity is forced to higher temperatures, as predicted by theory for the normal state (Makinson 1938, Sondheimer 1950). It is also seen that as the impurity increases the low temperature end of the superconducting curve is raised in relation to the normal curve. When we come to the specimen with 0·2% Bi the low temperature end of the superconducting curve actually lies above the normal curve, which it crosses at 4·8° K. At this impurity content the high temperature end of the curve still remains below K_n, but when the impurity is increased to 0·5% and 10%, K_s becomes greater than K_n throughout the range of measurement. The thermal conductivity of an alloy containing 30% of bismuth is shown in fig. 4 on an enlarged scale.

It is clear that these alloys behave quite differently from our pure specimens, and from all published results for pure metals. On the other hand the behaviour described here is not a general feature of high impurity content alloys. Thus an alloy of lead with 30% tin has a thermal conductivity in the superconducting state which is lower than that in the normal state throughout the range of measurement (Mendelssohn and Olsen 1950 a). The results shown in the same publication for a pure sample of niobium should not, however, be taken to indicate a real deviation from the usual characteristics of a pure superconductor. The magnetic field available during the measurements was not large enough to destroy superconductivity completely at the lower temperatures, and the curve shown as normal does not in fact represent the thermal conductivity with superconductivity destroyed. The crossing of the two curves in this case is therefore of little significance.

---

* The grain size was subsequently estimated as $\frac{1}{20}$ mm by etching a surface cut in the lead with a chisel. This seems to be surprisingly small, and although this method of preparing a surface is generally recommended, the result obtained thus must probably be accepted with a good deal of reserve.

(ii) *Discussion*

The thermal conductivity of a normal metal may be split up into two components, and we may conveniently write (Königsberger 1907)

$$K = K_e + K_g. \qquad \ldots\ldots(1)$$

Here K is the total thermal conductivity, K_e the electronic thermal conductivity, and K_g is the thermal conductivity due to the lattice vibrations. We shall denote by K_{en} and K_{es} the electronic and by K_{gn} and K_{gs} the lattice thermal conductivities in the normal and superconducting states respectively.

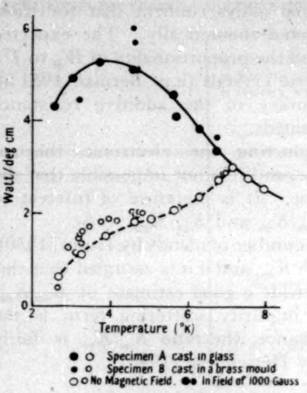


Fig. 2. Thermal conductivity of pure lead specimens.

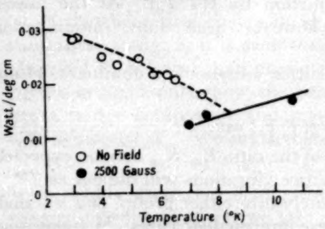


Fig 4. Thermal conductivity of alloy of lead with 30% bismuth.

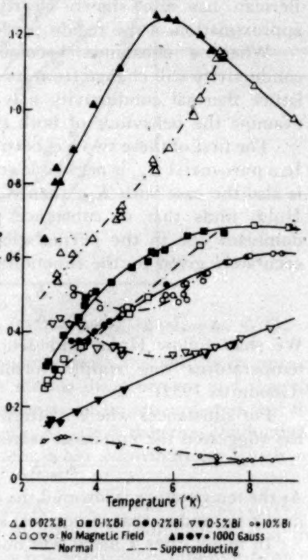


Fig. 3. Thermal conductivity of lead–bismuth alloys.

When we come to examine K_e and K_g separately it is simpler to consider their reciprocals. This is because the total thermal conductivity is made up of two conductivities in parallel, each of which is limited by resistances acting as if in series. In the normal state we have for the electronic contribution (Makinson 1938)

$$1/K_{en} = AT^2 + B/T, \qquad \ldots\ldots(2)$$

where the first term AT^2 is due to the scattering of the electrons by lattice vibrations with A a characteristic of the metal, and the second term is due to impurity scattering with B proportional to the residual electrical resistance.

For the lattice resistivity $1/K_g$, which will be denoted by W, the main contributions are: W_E due to the scattering by the electrons, W_B due to scattering at the boundaries of the specimen and crystal grains, and W_D due to scattering by impurities. We thus have

$$1/K_g = W = W_E + W_B + W_D. \qquad \ldots\ldots(3)$$

This simple sum may be used as a first approximation, but it cannot be relied upon implicitly. Klemens (1951) has pointed out that in dielectrics the corresponding expression becomes unreliable when two or more of the contributions to W are of the same order of magnitude. The experiments of Hulm (1950) and Berman (1951 a) on normal alloys confirm that for these W_E is proportional to T^{-2} as is to be expected theoretically. The expected linear dependence of W_D upon temperature, and the proportionality of W_B to T^{-3} have been verified in experiments on dielectric crystals (e.g. Berman 1951 b). Berman has also shown clearly the inaccuracy of the additive resistance approximation in the region predicted by Klemens.

When a substance becomes superconducting the electronic thermal conductivity will change from its normal value, and it is not impossible that the lattice thermal conductivity may do the same. It is therefore of interest to examine the behaviour of both the ratios K_{es}/K_{en} and K_{gs}/K_{gn}.

The first of these two has been found for a number of metals by Hulm (1950). In a pure metal K_{gn} is negligible compared with K_{en}, and if it is assumed that this is also the case with K_{gs}, then K_s/K_n will provide a good estimate of K_{es}/K_{en}. Hulm finds that in substances where the impurity scattering term is the dominant one in the normal electronic resistance, the ratio K_{es}/K_{en} is fairly accurately given by the function suggested by Heisenberg (1948)

$$\frac{K_{es}}{K_{en}} = \frac{2(T/T_c)^2}{1+(T/T_c)^4}. \qquad \ldots\ldots(4)$$

We shall follow Hulm and denote this function by $f(T/T_c)$. At the lowest temperatures this simple formula does however need some modification (Goodman 1951).

For substances where scattering by the lattice vibrations is dominant, Hulm has suggested the empirical relation

$$K_{es}/K_{en} = (T/T_c)^5 = g(T/T_c) \text{ say.} \qquad \ldots\ldots(5)$$

As the temperature is lowered the behaviour of the ratio K_{es}/K_{en} may be expected finally to tend to that of eqn. (4) since the lattice vibrations will die out as T^3.

In many cases a metal will not fall definitely into either group, and we shall need an interpolation formula to describe these intermediate cases. A suggestion for such an interpolation formula for K_{es} has been made by Hulm (1950), but it does not appear to give satisfactory agreement with the published results. We find that the experimental results are described slightly better by the formula

$$\frac{K_{es}}{K_{en}} = \frac{\alpha(T/T_c)^3 g(T/T_c) + f(T/T_c)}{\alpha(T/T_c)^3 + 1}. \qquad \ldots\ldots(6)$$

Where α is defined by $\alpha = AT_c^3/B$ so that it is the ratio of the two contributions to $1/K_{en}$ in (2) at the transition temperature. The formula (6) thus represents a mean of the two functions $f(T/T_c)$ and $g(T/T_c)$, weighted according to the amounts of the two scattering processes present at a particular temperature.

An examination of eqn. (2) shows that a plot of T/K_{en} against T^3 will be a straight line, and suggests an obvious way in which α may be evaluated. This has been done for some published experimental results, and in fig. 5 observed values for K_s/K_n for specimens with $\alpha = 0{\cdot}1$, 1 and 10 are compared with values of K_{es}/K_{en} calculated from eqn. (6). Agreement is seen to be fair in most cases at the higher temperatures. Hulm has shown how well the pure Heisenberg formula, to which (6) reduces for $\alpha = 0$, describes the experimental results for metal with high Debye θ where α is quite small even in very pure specimens. For the intermediate cases our calculation does at least give a better estimate of K_{es}/K_{en} than either $f(T/T_c)$ or $g(T/T_c)$ taken separately. At the lowest temperatures there is always a good deal of disagreement.

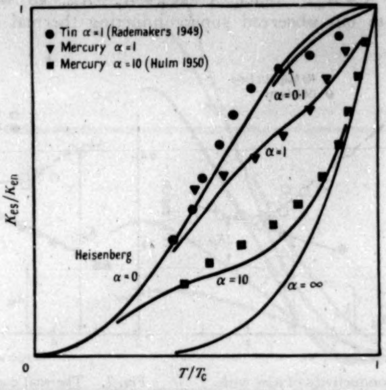


Fig. 5. K_{es}/K_{en} calculated from eqn. (6) compared with experimental values for K_s/K_n.

If the theoretical curves in fig. 5 are compared with our results in impure substances in fig. 3, it is seen that there is a serious disagreement and that the observed K_s is larger than predicted. It must however be remembered that the curves in fig. 5 only show the behaviour of K_{es}/K_{en}, and that the conductivity has a lattice component and may also contain other contributions which we will denote by X. We must therefore write

$$K_s = K_{es} + K_{gs} + X. \qquad \ldots\ldots(7)$$

It is readily seen that if K_{es} does really have the value predicted from the calculations above, and if we take it that K_{gs} is not appreciably different from its normal value, then X will represent an important part of the total conductivity.

It seems clear, of course, that the need for this extra term must be due to the fact that some important contribution has been omitted in calculating either K_{es} or K_{gs}. In fact formula (7) is very similar to one proposed in an earlier paper (Mendelssohn and Olsen 1950 a) in which the extra term was considered to be part of the electronic conductivity in the form of a circulation process similar to that in liquid helium II. It was then pointed out that while this process could be made to fit our observed data, numerical evaluation would be unconvincing because of the difficulty of checking the assumptions made for this process. It is, of course, also possible that the high value of K_s may be due to some increase in the lattice conductivity in the superconducting state. Indeed Hulm (1950) has pointed out that a reduced scattering of lattice waves by the superconducting electrons may cause an enhanced thermal conductivity of the lattice. Since the proportion of X or K_{gs} in such substances as our alloy of lead with 10% bismuth is larger by an order of magnitude than found in any of Hulm's results, explanation by an enhanced lattice conductivity did not appear convincing at first sight. However, the results on the graded series of lead–bismuth alloys described in the present paper offer an opportunity of estimating the postulated enhanced lattice conductivity in a more quantitative manner.

The most convenient first step in our estimate of the magnitude of X will be to calculate $X+K_{gs}$. Since $X+K_{gs}=K_s-K_{es}$, all we need do is to subtract K_{es} from our observed superconducting thermal conductivity. We have used formula (6) to derive K_{es} from the normal electronic thermal conductivity K_{en}. The values of α required were found as described above and ranged from very nearly zero for the most impure specimens, to $\alpha=2$ for the specimen containing 0·02% bismuth. In principle K_{en} may be derived from the electrical conductivity and a knowledge of the appropriate value of the Wiedemann–Franz constant. Figure 6 shows how we have used this procedure to obtain K_{es} for the specimen containing 10% bismuth.

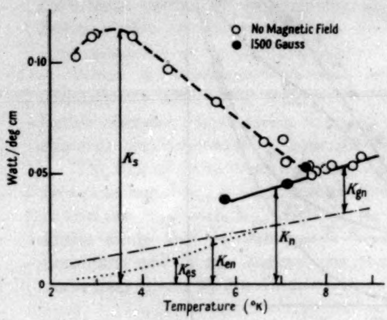


Fig. 6. Thermal conductivity of lead with 10% bismuth.

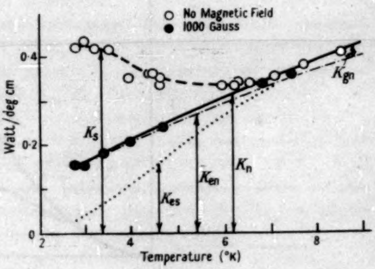


Fig. 7. Thermal conductivity of lead with 0·5% bismuth.

This simple method of calculating K_{en} cannot be used with the purer specimens, because the electrical data are not sufficiently accurate, and because the Wiedemann–Franz constant is not reliably known for them. In order to calculate K_{en} we have therefore subtracted a calculated value of K_{gn} from the total normal thermal conductivity. This value for K_{gn} has been derived from the lattice conductivity of the 10% bismuth specimen between 7° K and 21° K. In this region the only important factors limiting K_{gn} are electron and impurity scattering, and a knowledge of their theoretical dependence on temperature allows them to be separated. We thus obtain W_{En} and W_{Dn} for the impure specimen. In the purer specimens the impurity scattering will be negligible compared with the scattering by electrons, and it has been assumed that the resistance from this cause, $W_{\rm En}$, is the same in the pure and the impure specimens. The value thus obtained for $W_{\rm En}$ in lead is 25 deg cm/watt at 10° K. This can be compared with the value of 50 deg cm/watt to be expected from the expression given by Makinson (1938). We would here remark how much better is the fit of this expression for the lattice thermal resistance in the presence of electrons, than is the fit of the expression given in the same publication for the electronic thermal resistance in the presence of lattice vibrations.

In fig. 7 is shown the conductivity of the specimen containing 0·5% Bi. $K_{\rm en}$ has been calculated by subtracting from the total normal thermal conductivity the value for $K_{\rm gn}$ described above.

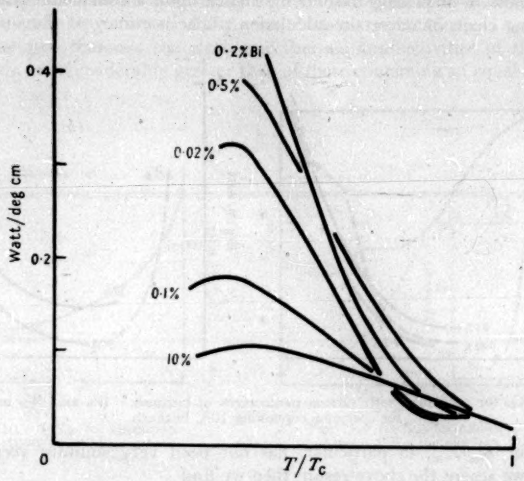


Fig. 8. Contribution to $K_{\rm s}$ other than $K_{\rm es}$ for specimens with various percentages of bismuth.

In fig. 8 are given the differences between $K_{\rm es}$ and $K_{\rm s}$ for the whole series of lead bismuth specimens with the exception of the two pure specimens for which $K_{\rm s} - K_{\rm es}$ is too small a fraction of $K_{\rm s}$ for reliability, and the 30% Bi specimen which belongs to a different crystal system. The fact that all the curves coincide at $T = T_{\rm c}$ is simply a consequence of the way in which we have calculated the lattice thermal conductivity in the normal state. All the curves rise with falling temperature, and appear to reach a maximum at about $0{\cdot}4\,T_{\rm c}$. Assuming tentatively that X is zero and that $K_{\rm s} - K_{\rm es}$ is indeed to be interpreted as $K_{\rm gs}$, we have for more convenient discussion plotted in fig. 9(*a*) and (*b*), instead of $K_{\rm gs}$, $W = 1/K_{\rm gs}$ against $T/T_{\rm c}$. The resultant curves for all five specimens are seen to be very much alike.

In fig. 9(*b*) we have also shown the terms $W_{\rm B}$ and $W_{\rm D}$ due to scattering at grain boundaries and impurities calculated for the 10% Bi specimen in the normal state. A comparison of the total observed W with these two contributions shows that they are of the same order of magnitude at the lowest temperature of measurement. This would seem to indicate that the only other contribution to W, the scattering of the lattice vibrations by electrons, should be negligible at this temperature. Thus the high values of the observed thermal conductivity might indeed be explained by a rapid decrease of the scattering of the lattice vibrations by the electrons in the superconducting state. If we consider such a process possible it is of course tempting to try to derive the temperature variation of the scattering of phonons by electrons in the superconducting metal.

Assuming this to be the same function for all the specimens we obtain $W_{Es} \propto (T/T_c)^n$, where n is about 4. When we take into account the uncertain validity of eqn. (3) in the region of the minima in K_g then the results are probably in agreement with this power law to within the limits of error. We should certainly beware of placing too much reliance upon a conclusion reached after such a long chain of uncertain calculation. The accuracy of the interpolation formula for K_{es}/K_{en}, in particular, has not been very soundly verified. If, however, we accept the above result, then we find

$$W_{Es}/W_{En} = (T/T_c)^6 \qquad \ldots\ldots(8)$$

which we may compare with the fifth-power law for K_{es}/K_{en} suggested by Hulm (1950) for the case when the electrons are scattered only by lattice vibrations.

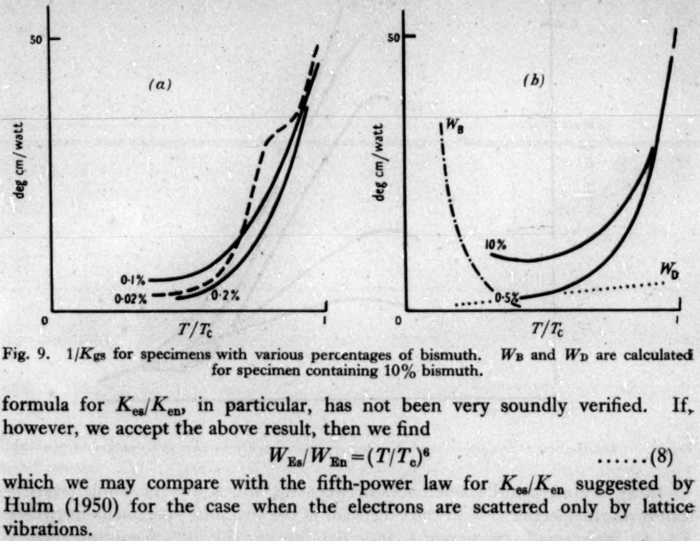


Fig. 9. $1/K_{gs}$ for specimens with various percentages of bismuth. W_B and W_D are calculated for specimen containing 10% bismuth.

The lattice resistivity W of the specimen containing 30% bismuth is shown in fig. 10. In considering this diagram it must be borne in mind that this alloy belongs to a different crystal system from that of the other lead–bismuth alloys examined. In addition we must remember that the amount of impurity scattering will be greater than in these specimens. With these allowances the behaviour appears in at least qualitative agreement with the explanations given in the previous section.

(iii) *Comparison with other Results*

It is of interest to see if the picture given above and deliberately fitted to our results for the lead–bismuth alloys with bismuth contents below 10% will fit other results as well.

The previously published (Mendelssohn and Olsen 1950 a) thermal conductivity results for niobium did not contain reliable values for the normal conductivity, but these may be deduced from electrical measurements. A value for K_s/K_{en} derived from these data is shown in fig. 11, and the corresponding lattice resistivity is shown in inset. The agreement with the treatment given in the previous section is only partly satisfactory. An extrapolation of the lattice resistivity curve according to a T^4 law would lead to a value of 1 000 deg cm/watt for the lattice resistivity at the transition temperature, and this appears to be approximately ten times greater than the value to be expected on the basis of Makinson's (1938) theory.

The fact that the alloy containing 30% tin does not behave as the bismuth alloys and does not have an abnormally high value for the thermal conductivity in the superconducting state naturally requires some explanation. There would appear to be two reasons: the electronic thermal conductivities of the alloys containing tin are considerably greater than of those containing an equal impurity of bismuth which belongs to a different group in the periodic table; in addition to this, while the electrons are less affected by the presence of tin than of bismuth, one may expect that because of the greater difference in atomic mass the lattice vibrations will be more strongly scattered by tin than by bismuth impurity. The situation thus arises in this alloy, that although only a fairly large increase in K_g would be noticeable, there is too great a resistance due to impurity scattering of the lattice vibrations to allow K_{gs} to be increased much above K_{gn}. To make these reasons completely convincing, however, it is clearly desirable to have much more information on the heat conduction in other superconductive alloy systems.

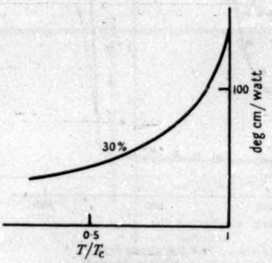


Fig. 10. $1/K_{gs}$ for specimen containing 30% bismuth.

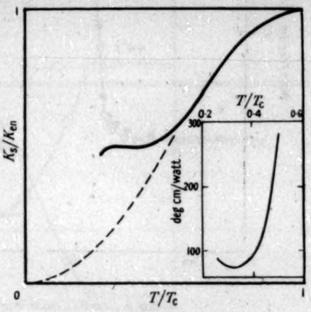


Fig. 11. K_s/K_{en} for niobium. Inset: $1/K_{gs}$ for niobium.

An examination of the results obtained by Hulm (1950) for the lattice conductivity in superconducting tantalum, and in tin of high impurity content appears to indicate that perhaps the fourth-power variation with temperature of K_{gs} which we have deduced for lead is not a general feature; or else, and perhaps more probably, that we have obtained too high a power for the temperature dependence of K_{gs}.

§4. FIELD TRANSITIONS

Further information on the conduction mechanism can be gained by measuring the thermal conductivities in the interval of temperature and magnetic fields in which the actual destruction of superconductivity takes place (Mendelssohn and Pontius 1937). Such measurements have been carried out in longitudinal and transverse fields for the alloys of the lead–bismuth series.

In fig. 12 are shown conductivities of our two purest specimens in a longitudinal field. It will be seen that just as the transitions measured by Mendelssohn and Pontius (1937) on lead, and by Hulm (1950) on tin, these are fairly sharp, and that any lack of sharpness may be accounted for by the demagnetization factor of the specimens, which are not infinitely long. The observed width of the transition at 4·6° K is somewhat greater than the width of the electrical transitions found by MacDonald and Mendelssohn (1949) in long specimens at the same temperature. At lower temperatures where these authors found very wide electrical transitions, the thermal conductivity changes as sharply from its superconducting to its normal value as at the higher temperatures. For comparison we have also shown the shape of a transition of the electrical resistance ρ in the diagram for 2·7° K. It is plotted so that $\rho=0$ coincides with K_s and $\rho=\rho_{\text{normal}}$ coincides with K_n. The greater width of the electrical transition, and the fact that the transition in the thermal conductivity remains sharp just as the magnetic transition, would appear to lend support to the view that the wide electrical transitions observed in lead are due to the existence of only very small regions having high critical field.

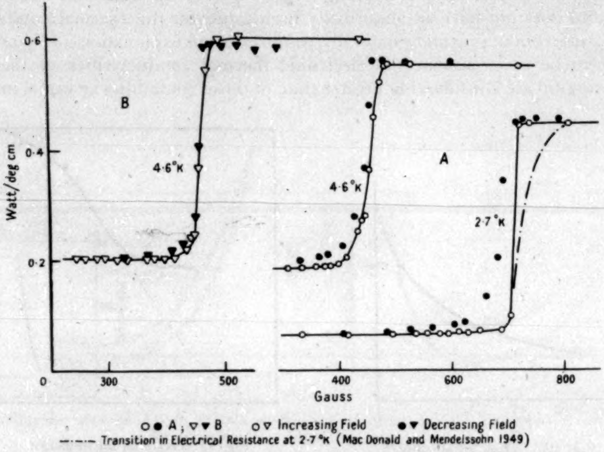


Fig. 12. Thermal conductivity in longitudinal magnetic field for pure lead specimens.

In fig. 13 we show the transition of a pure specimen in a transverse field at 3·3° K. We see that the transition in conductivity is far from linear. If the same transition is plotted in terms of thermal resistance, on the other hand, it is found that the variation is almost linear with field. A very simple calculation shows that this is an indication that the laminae forming the intermediate state (Landau 1943) lie at right angles to the axis of the cylindrical specimen. The argument leading to this result is as follows: If the fraction of the volume of the cylinder occupied by normal and superconducting material are a and b respectively, then the conductivity of the cylinder will be given for the case of longitudinal laminae by

$$K=(aK_n+bK_s)/(a+b) \qquad \ldots\ldots(9)$$

but if the laminae lie perpendicular to the axis of the cylinder, then

$$1/K=(a/K_n+b/K_s)/(a+b). \qquad \ldots\ldots(10)$$

It is known from magnetic experiments (Shoenberg 1948, etc.) that once $\frac{1}{2}H_c$ has been exceeded for a cylinder in a transverse field, then the amount of normal material increases approximately linearly with field until all the material is normal at H_c.

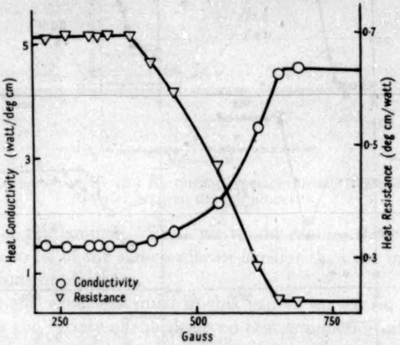


Fig. 13. Transverse field transition at 3·3° K for pure lead specimen B.

An anomalous effect indicating a rather more complicated behaviour has been found in specimens with a small degree of impurity or strain (Mendelssohn and Olsen 1950 a, c). Similar results have recently also been obtained on pure lead, particularly at low temperatures and under complicated geometrical conditions (Webber and Spohr 1951).

In fig. 14 is shown the variation of thermal conductivity with field for our specimens of intermediate purity. At the highest temperatures there is a transition which is very like that observed in pure specimens. The departure from linearity is less obvious than in these, but this is simply a consequence of the much lower ratio of K_s to K_n. At a somewhat lower temperature there is a sharp drop in conductivity when $\frac{1}{2}H_c$ is exceeded, and the conductivity then recovers and rises to its value in the normal state. At a still lower temperature only the initial drop is found, and the recovery is almost non-existent. The return curves in a decreasing field all lie below the increasing field curves.

This anomalous drop in the conductivity between the completely normal and the completely superconducting state creates a difficulty when an explanation is attempted. As we have already pointed out when announcing the first of these results, it is impossible to interpret the results simply as being due to a set of successive laminae of alternating heat conductivity. A series of thermal resistances of which the individual components have either the normal or the superconductive resistance must clearly always result in an overall value intermediate between these two resistivities, as for example in the case in fig. 13. A heat conduction which is lower than in either of these states cannot be explained in this simple way, but requires some more complicated mechanism, such as for example the circulation process referred to previously.

The only other explanation for the heat conduction results in superconductive alloys is that of the enhanced lattice conductivity, and it is therefore interesting to see what additional assumption would have to be made for this alone to explain the experimental results. It is interesting to note that by calculating the lattice and electronic heat conductivities separately, and treating them as parallel throughout the length of the specimen, a minimum in the heat conductivity in the intermediate state can be produced. This is equivalent to assuming that the two heat currents flow independently without transfer from one to the other. From what has been said in the preceding section we should expect the overall K_g to decrease rapidly as superconductivity is destroyed, while at the same time

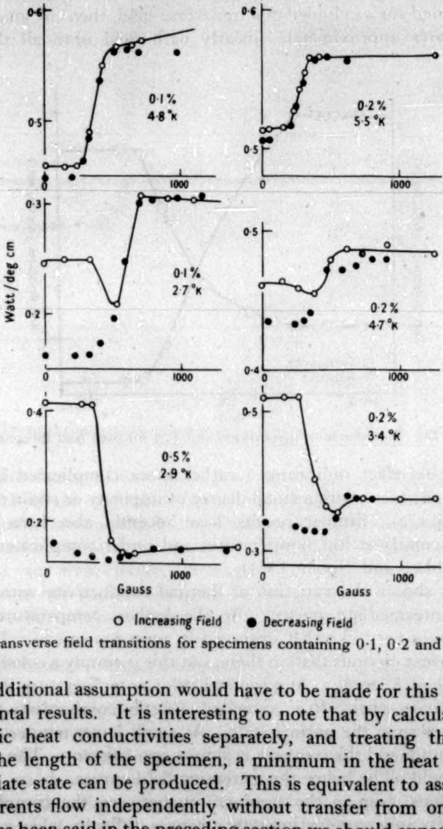


Fig. 14. Transverse field transitions for specimens containing 0·1, 0·2 and 0·5% bismuth.

K_e increases. Little can be said, of course, concerning the way in which these changes depend on the field, but as fig. 15 shows a minimum can be produced by a suitable adjustment of the constants.

The trouble with this picture is that it appears to contradict our concept of an intermediate state made up of a set of laminae each with their own value of the thermal conductivity. However, it serves as a useful guide to show what assumptions would have to be made to explain the phenomena by an enhanced lattice conduction. It seems clear that in order to obtain an approach to the case of parallel conduction processes (a concept which is incidentally similar to the circulation hypothesis) there must be little interchange of energy between the lattice and the electronic heat currents over a distance of the order of magnitude of the thickness of the laminae. Thus we would require the periodicity of the laminar structure to be of the same order or smaller than the mean free path of the lattice vibrations or electrons.

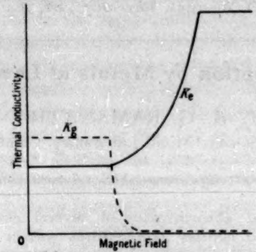


Fig. 15. Expected variation of K_e and K_g during superconducting transition with zero energy transfer between the two processes.

Summarizing the work described in this paper, we see no easy and simple way of explaining the variety of the observed heat conduction phenomena. The validity of such ideas as a circulation process or the enhanced lattice conductivity must still rely on assumptions which can only be checked by further experiments. The present work shows that the second as well as the first of these processes might be capable of explaining the phenomena, and that so far neither process can be ruled out by the evidence provided. Beside the recording of the anomalous phenomena associated with heat flow in superconductive alloys, experiments of this kind may offer a useful new method for investigating the intermediate state.

ACKNOWLEDGMENTS

It is a very great pleasure to thank Dr. K. Mendelssohn for suggesting this investigation and for his constant encouragement and help. The author is much indebted to Dr. J. W. Christian for taking photographs of the grain structure of some of the specimens. This work was carried out during the tenure of a grant from the Department of Scientific and Industrial Research.

REFERENCES

BERMAN, R., 1951 a, *Phil. Mag.*, **42**, 642; 1951 b, *Proc. Roy. Soc.* A, **208**, 90.
GOODMAN, B., 1951, International Low Temperature Conference Report, Oxford.
DE HAAS, W. J., and RADEMAKERS, A., 1940, *Physica*, **7**, 992.
HEISENBERG, W., 1948, *Z. Naturforsch.*, **3a**, 65.

HULM, J. K., 1950, *Proc. Roy. Soc.* A, **204,** 98; 1951, *Proc. Phys. Soc.* B, **64,** 207.
KLEMENS, P. G., 1951, *Proc. Roy. Soc.* A, **208,** 108.
KÖNIGSBERGER, J., 1907, *Phys. Z.*, **8,** 237.
LANDAU, L., 1943, *J. Phys., U.S.S.R.*, **7,** 99.
MAKINSON, R. E. B., 1938, *Proc. Camb. Phil. Soc.*, **34,** 474.
MACDONALD, D. K. C., and MENDELSSOHN, K., 1949, *Proc. Roy. Soc.* A, **200,** 66; 1950, *Ibid.*, **202,** 103.
MENDELSSOHN, K., and OLSEN, J. L., 1950 a, *Proc. Phys. Soc.* A, **63,** 2; 1950 b, *Ibid.*, **63,** 1182; 1950 c, *Phys. Rev.*, **80,** 859.
MENDELSSOHN, K., and PONTIUS, R. B., 1937, *Phil. Mag.*, **44,** 777.
RADEMAKERS, A., 1949, *Physica*, **15,** 849.
SHOENBERG, D., 1948, *Proc. Roy. Soc.* A, **197,** 65.
SIMON, F. E., 1932, *Z. f. Kälteindustrie*, **39,** 89.
SONDHEIMER, E. H., 1950, *Proc. Roy. Soc.* A, **203,** 75.
WEBBER, R. T., and SPOHR, D. A., 1951, *Phys. Rev.*, **84,** 384.

Infra-Red Absorption by Metals at Low Temperatures

BY K. G. RAMANATHAN
Royal Society Mond Laboratory, Cambridge

Communicated by D. Shoenberg; MS. received 19*th February* 1952

ABSTRACT. The infra-red absorptivities of several electropolished metal surfaces have been measured at liquid helium temperatures with a specially designed radiation cryostat with a black body at room temperature as the source and a differential helium gas thermometer as the detector. It is found that the absorptivity of tin at 1·95° K for the mean estimated wavelength 14 μ remains the same in the normal and superconducting states to within a probable error of 0·3%. The absorptivities of the normal metals, which include a number of alloys, are compared with the values calculated by various theories of metallic reflection. The classical Hagen–Rubens formula is found to be valid for the alloys except where there are complications such as the internal photoelectric effect. The absorptivities of the pure metals are in fair agreement with the values calculated by the anomalous skin effect theory neglecting relaxation effects.

§ 1. INTRODUCTION

THE classical electromagnetic theory of light leads to an expression for the absorptivity A and reflectivity R of a metal surface at normal incidence

$$A=(1-R)=2(\nu/\sigma)^{1/2}, \qquad \ldots\ldots(1)$$

where ν is the frequency of the radiation and σ the d.c. conductivity of the metal in electrostatic units. This suggests the interesting possibility that superconducting metals might be particularly good reflectors of high-frequency radiation. In the infra-red region, Daunt, Keeley and Mendelssohn (1937) carried out absorption measurements on lead and tin using the radiation from a black body at room temperature as the source. Estimates of the relative absorption in the normal and superconducting states of the metal were made by measuring the rates of evaporation of liquid helium contained in an attached calorimeter. From these observations it was concluded that the transition to superconductivity does not affect the infra-red absorptivity of a metal. Experiments in the microwave region were first made by H. London (1940) who showed that the absorptivity of a metal, when cooled through its transition point, does not fall suddenly to zero as indicated by (1) but rather rapidly at first and then more gradually, apparently tending to zero at the absolute zero of temperature. More recently Pippard (1947 a, 1950) has used a resonance technique to carry out extensive investigations of this phenomenon at 1 200 and 9 400 Mc/s, and has confirmed the results of London by this independent method. These results have, during recent years, aroused considerable interest in the behaviour of superconductors at still higher frequencies and also made it desirable to approach the problem from the infra-red end of the spectrum. The scope of the investigation at infra-red frequencies is at present necessarily limited to the shorter wavelengths owing to the lack of monochromatic sources of adequate intensity for wavelengths longer than about $20\,\mu$. However, extrapolation from the work of Pippard suggests that even at such short wavelengths as $10\,\mu$, the absorptivity of a metal might show a small decrease of the order of a few per cent in the superconducting state. Since the work of Daunt, Keeley and Mendelssohn did not claim a high degree of accuracy, it was decided to make new measurements on the infra-red absorption by tin with an apparatus designed to give results accurate to better than 1%.

The high-frequency study of metals, besides throwing light on the phenomenon of superconductivity, has also led to interesting results about the behaviour of normal metals. During his investigation of the behaviour of tin in the microwave region, H. London observed that the surface resistivity of the metal above its superconducting transition point was higher than that predicted by the classical theory. He attributed the anomaly to the fact that at low temperatures the mean free path of the conduction electrons is either comparable with or much larger than the depth of penetration of the electric field into the metal. This observation by London has also been confirmed by Pippard's (1947 b) measurement of surface resistivities of copper, silver, gold, aluminium, tin and mercury and particularly by the later detailed study by Chambers (1950 a) of copper and silver. From simple theoretical considerations Pippard predicted that the surface resistivity of metals at microwave frequencies should vary as the two-thirds power of the frequency. The detailed theory, taking account of the variation of the electric field along the path of conduction electrons, has been worked out by Reuter and Sondheimer (1948) using the general methods of the theory of metals. The results of this theory seem to offer a satisfactory explanation of all observed data on normal metals in the microwave region. The verification of the predictions of the theory regarding the behaviour of metals in the infra-red region has not so far been possible due to lack of experimental data. Therefore it was also the aim of this investigation to measure quantitatively the absorptivities of some pure metals and alloys and compare the results with those predicted by various theories, including that of Reuter and Sondheimer.

.A
5
2

§ 2. EXPERIMENTAL TECHNIQUES

(i) *Method of Investigation*

In the present method, as in that of Daunt, Keeley and Mendelssohn, the absorption by the specimen of radiation from a source at room temperature is studied. The radiation from the source passes down the cryostat through a German silver tube T (fig. 1), polished on the inside to increase the effective intensity by multiple reflections at the wall, and falls on the flat polished surface of the metal specimen S under investigation. The energy absorbed is measured by the heating of one of two identical gas thermometer bulbs to which the specimen is soldered and which are enclosed in a highly evacuated metal chamber. The gas thermometers are connected by metal capillaries to either side of a miniature oil manometer at the top of the cryostat. The lower portions of the tube T, the capillaries and the metal chamber are immersed in liquid helium during an experiment. When the shutter Sh (which is at a low temperature and therefore does not itself radiate appreciably) is opened, radiation falls on the specimen and the increase in pressure in the thermometer bulb to which it is attached causes a displacement of the levels in the oil manometer, which is measured by a travelling microscope. The power absorbed by the surface is obtained by comparing the change in oil level caused by the radiation with that produced by dissipating a known electrical power in a heating coil H wound round the gas thermometer carrying the specimen. In this way knowledge of the absolute temperature rise is unnecessary, and various correction factors are eliminated. In order to observe as accurately as possible whether there is a change in absorption when the specimen changes from the superconducting to the normal state, the cryostat was surrounded by a pair of Helmholtz coils which could give a magnetic field sufficient to destroy superconductivity.

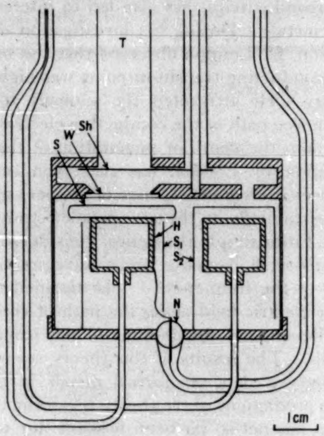


Fig. 1. Experimental arrangement.

The effective intensity of the source, which is required to calculate the absolute absorptivities of the various metals investigated, was estimated by measuring the absorption by a surface thickly coated with zinc black powder, known to absorb the incident radiation almost completely in the spectral region in question. The value thus obtained was checked independently by measurements on alloys whose absorptivities could be estimated from the classical formula (1).

(ii) *Experimental Details*

The factors considered in designing the gas thermometer were its rise in temperature above that of the helium bath, the time taken to attain equilibrium when exposed to radiation and the effect of dead space on the sensitivity. The rise in temperature is proportional to the thermal resistance of the capillary connecting it to the bath and the incident radiant flux which is independent of the detector itself. The equilibrium time is proportional to the thermal resistance of the capillary and to the total heat capacity of the thermometer (including the specimen and gas), which was kept as low as possible since it does not influence the sensitivity. The capillaries were made of an 80 : 20 cupro-nickel alloy with dimensions chosen to give a reasonably short equilibrium time (about 3 minutes) and a sensitivity of the order of 10^{-6} watt per mm of oil at 4·2° K for a gas pressure of a few cm Hg. The narrow capillaries (0·5 mm) used at first to minimize the dead space were frequently blocked by impurities in the helium and so had to be replaced by wider ones (1·5 mm), in spite of the consequent reduction of sensitivity by about 50%. It was not possible to use the gas thermometer below about 2° K, since the rapidly falling vapour pressure restricts the working pressure too greatly and makes the thermometer too insensitive.

The metal shields S_1 and S_2, soldered to the bottom of the vacuum chamber, prevent scattered radiation from falling on the sides of the thermometers. These shields decreased the apparent absorption from about 350 to 140 erg/sec, but introduction of additional shields carefully designed to eliminate the small influx of radiation through the gap below the specimen caused no further improvement. The consistency of the results obtained with different specimens of the same metal was taken as evidence that the shields S_1 and S_2 were performing their task efficiently. The thin ($\frac{1}{2}$ mm) silver chloride* window W, fixed above the specimen, is used to prevent 'hot' molecules of the residual gas from heating up the thermometer on opening the shutter. One end of the heating coil is soldered to the gas thermometer while the other end is taken out through a glass–metal seal N, and then through a thin-walled German silver tube in order to shield it from extraneous high-frequency radiations such as that from a neighbouring cyclotron. A high vacuum was maintained in the chamber during the experiments by first evacuating it at room temperature with an oil diffusion pump, then isolating it from the pump by a tap and relying on cooling in liquid helium to freeze out the residual gases.

(iii) *Specimens*

The specimens listed in table 1 were made as circular discs of about 3 mm thickness and $\frac{7}{8}$ in. diameter. Their electrolytic polishing was carried out by well-known techniques (Kemsley 1948). They were all soldered with Wood's metal symmetrically to the thermometer. The electrical resistivities of the specimens (which are needed in the theoretical discussion) were, for the alloys, obtained from measurements on strips cut out of the specimens after the absorption measurements were finished and, for the pure metals, were taken from published data.

(iv) *Calibration and Performance*

To test the constancy of the source of radiation, the absorption by a copper surface was measured in the range from 4·2° to 2° K. Since theoretically there is no obvious reason why the absorption should vary, any apparent variations must be attributed to variations of the effective intensity of the source. The result of this experiment, illustrated in fig. 2, indicates a gradual increase in absorption of about 6 or 7%. Detailed examination of this variation in terms of the level of helium in the bath and the rate of pumping suggested that the effective intensity depends in a complicated way on the distribution of temperature in the top portions of the radiation tube and therefore is not exactly reproducible from run to run. When the absorption was not expected to vary with temperature, the mean of all the readings in any one experiment was taken as the appropriate value and this should be accurate to about 3%. This was confirmed by the consistency of results obtained on the same specimen in different runs.

---

* At room temperature the transparency of a thin silver chloride window is known to be uniformly about 70% in the range 1 to 40 μ; at low temperatures the transparencies of solids in general increase and extend to longer wavelengths.

Table 1. List of specimens

Specimen	Method of preparation
Electrolytically pure copper	Machined from rod, annealed and electropolished.
Electrolytically pure copper	As above but polished with emery paper then with 'Brasso' instead of electropolishing.
Tin J.M. 2356 ($<0{\cdot}004\%$ impurity)	Cast against glass and electropolished.
Lead J.M. 560 ($<0{\cdot}005\%$ impurity)	Cast against glass and electropolished.
Aluminium from British Aluminium Company ($<0{\cdot}01\%$ impurity)	Cut from sheet, annealed and electropolished.
Tin J.M. 2356 alloyed with 1% and with 5·4% chemically pure indium	Cast against glass and electropolished.
57% Kahlbaum bismuth + 43% chemically pure tin	Cast against glass.
Brass Eureka and Staybrite } commercial stock	Cut from sheets, ground with sand paper and electropolished.

Note: J.M. = Johnson Matthey.

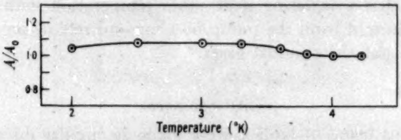


Fig. 2. Variation of apparent absorption A by copper surface expressed as a ratio A/A_0 where A_0 is the lowest observed value.

The absolute value of the effective intensity of the source, which was required to estimate the absorptivities of metal surfaces, was determined by measuring the power absorbed by each of two surfaces, one blackened with zinc black and the other with Aquadag; these were found to be 11 000 erg/sec and 8 000 erg/sec respectively. Since at room temperature zinc black is known to absorb almost completely and Aquadag from 60 to 70% of the incident radiation in the infra-red region under consideration, these measurements may be regarded as reasonably consistent and the incident power was assumed to be 11 000 erg/sec. Taking into account the variation of intensity during each experiment, this estimate of the effective intensity is probably accurate to only about 3% and the final absorptivities quoted in column (3) of table 2 to only about 5%.

The mean wavelength to which the absorption measurements refer was deduced using Planck's formula for the spectral distribution of intensity in black body radiation. If the incident intensity is given by $\int_0^\infty E_\nu \, d\nu$, the total energy absorbed by the metal surface can be represented by

$$\int_0^\infty c\nu^a E_\nu \, d\nu,$$

where c and a are independent of the frequency ν. The experimentally measured absorptivity is the quotient

$$\int_0^\infty c\nu^a E_\nu \, d\nu \bigg/ \int_0^\infty E_\nu \, d\nu$$

which may be regarded as the absorptivity $c\nu_1{}^a$ at some mean frequency ν_1. It is found by integration that the corresponding mean wavelength λ_1 does not vary appreciably with a, being $14{\cdot}0\,\mu$ when $a = \frac{1}{2}$ and $13{\cdot}6\,\mu$ when $a = \frac{2}{3}$.

§ 3. EXPERIMENTAL RESULTS AND DISCUSSION

(i) *Superconductors*

Although a number of superconductors such as tin, lead and alloys of tin with indium and bismuth were investigated, attention was mainly focused on tin. The absorption measurements on tin in the range 4·2° to 2° K do not indicate any change corresponding to the transition to superconductivity at 3·7° K but only the gradual apparent increase in absorption which, as with the copper surface, could be attributed to an increase of the effective intensity of the source. Thus this experiment indicated only that the absorption did not vary by more than about 5 % down to 2° K. To achieve the higher accuracy permitted by the detector the cryostat was cooled to a temperature (1·95° K) at which the detector still works satisfactorily and the absorption was measured at regular intervals of time alternately in the superconducting state and in the normal state produced with the aid of a magnetic field. As can be seen from fig. 3 the two sets of readings corresponding to the normal and superconducting states plotted against time lie on the same curve within the accuracy of the detector, indicating that the absorptivity of tin is the same in both states. The probable uncertainty in the constancy of absorption was estimated as less than 0·3 % from the deviations of one of the two sets of points from a smooth curve drawn through the other.

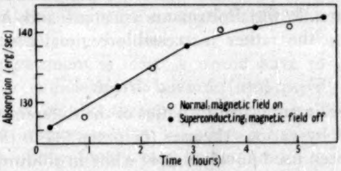


Fig. 3. Absorption by a tin surface in the normal and superconducting states at different times.

During the experiments it was noticed that switching the magnetic field on and off, besides causing an eddy current heating of the thermometer (and the specimen in the normal state) which gave rise to a kick in the oil levels of about 1 mm, produced a kick of about 1 cm, caused by the latent heat accompanying the transition of the specimen from the normal to the superconducting state or vice versa. The points shown in fig. 3 were obtained after waiting the necessary time for thermal equilibrium to be restored.

In the case of the other superconductors the magnetic method was not used and hence the constancy of their absorption can be inferred only to within about 5%. For lead this is not particularly significant since the temperature range investigated is well below the transition point 7·2° K and no large change with temperature was to be expected; for the alloys, however, an appreciable change might have been expected because of their high residual resistance in the normal state.

Using dimensional arguments Pippard (1949) deduced that just below the transition temperature the gradient dr/dT in the resistivity–temperature curve should vary as $\nu^{-2/3}$($r = R/R_n$ where R and R_n are the surface resistivities in the superconducting and normal states respectively) and this was confirmed by observations on tin at the two frequencies 9 400 and 1 200 Mc/s. At 9 400 Mc/s dr/dT at the transition temperature is about 20 and it should therefore be about 0·12 for the infra-red frequency used, if extrapolation according to the $\nu^{-2/3}$ law is permissible over such a wide range. Even though of course dr/dT becomes smaller as the temperature falls below the transition temperature, this would suggest that a change of a few per cent in r and therefore in absorptivity might be expected when the specimen becomes superconducting at a low temperature. The reason why such a change is not in fact observed is probably because the theoretical argument is based on the 'two-fluid' model which assumes the coexistence of 'normal' and 'superconducting' electrons. At these high frequencies relaxation effects in the normal electrons should probably also be considered, but this would lead to an even more pronounced fall of resistance in the superconducting transition. However the two-fluid model already disagrees with some aspects of Pippard's observations at 9 400 Mc/s and might be expected to break down altogether at still higher frequencies due to the setting in of new absorption mechanisms. It is possible that the frequency at which such mechanisms set in is of order of magnitude $\boldsymbol{k}T_c/\boldsymbol{h}$ (where T_c is the transition temperature, $\boldsymbol{k}$ the Boltzmann constant and $\boldsymbol{h}$ Planck's constant) which corresponds to the rather inaccessible region of wavelengths of a few millimetres.

(ii) *Normal Metals*

In table 2 the experimental absorptivities of the different metals are compared with those calculated by various theories (columns (4) to (8)). The formula of classical theory has been used in column (4) while in column (5) the absorptivity is assumed to be given by $2(\nu/\sigma)^{1/2}\{-\omega\tau+(1+\omega^2\tau^2)^{1/2}\}^{1/2}$ where $\omega = 2\pi\nu$ and τ the 'relaxation' time is equal to $\sigma\boldsymbol{m}/n\boldsymbol{e}^2$, n being the number of conduction electrons per cm^3, $\boldsymbol{e}$ the electronic charge and $\boldsymbol{m}$ its mass. The calculations in columns (6) to (8) are based on the theory of the anomalous skin effect in metals developed by Reuter and Sondheimer. The parameter p in these columns is the fraction of the conduction electrons reflected specularly on collision with the metal surface. Chambers (1950 b) has shown that his results on the electrical resistivities of thin wires are in agreement with a value of $p = 0$. For the optical region Reuter and Sondheimer have made their computations only for $p = 1$. The values in columns (6) and (7) are based on fig. 1 of their paper which represents their results if relaxation effects are assumed negligible. The values in column (8) are obtained by interpolation from fig. 2 of their paper which represents their results for the optical region where relaxation effects play an important part in determining the surface resistance. In all these calculations, the number of free electrons per atom has been assumed to be 1; the numerical values are not much changed if n is changed by a factor of 2. It may be otend that these assumptions if used to calculate the resistivities at microwave frequencies give fair agreement with the available experimental data (Pippard 1947 b, Chambers 1950 a, Maxwell, Marcus and Slater 1949), except where a poor surface was used.

It can be seen that the calculated absorptivities assuming $p=0$ and no relaxation effects (column (6)) are in fair agreement with experimental values while those in other columns, for most cases, lead to absorptions lower than observed, this discrepancy being more marked in the case of the pure metals and the 1% indium alloy for which the resistivities are low enough to produce the anomalous skin effect. These features suggest that relaxation is unimportant even at $14\,\mu$ in all these metals except perhaps for copper which approximates better than the others to the free electron model on which the theory is based. That the values in the last column are not appreciably influenced by the value of n chosen can be inferred from the fact that, even for copper for which the disagreement with experiment is least, n would have to be increased five or six times to reduce τ sufficiently to give an absorptivity in agreement with experiment. Extension of the theory to the case $p=0$ might explain why relaxation effects are unimportant at $14\,\mu$, but it is possible that some more fundamental revision of the theory may be required.

Table 2. Experimental and theoretical percentage absorptivities at $\lambda=14{\cdot}0\,\mu$.

(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)
				Theoretical			
Metal	d.c. resistivity $\times 10^6$ ohm × cm at 4·2° K	Experi-mental	Classical theory	Classical + relaxation	Anomalous $p=0$	Skin effect $p=1$	Anomalous + relaxation
Copper	0·011	0·62	0·10	0·003	0·94	0·84	0·1
Copper (M.P.)	0·011	1·47	0·10	0·003	0·94	0·84	0·1
Aluminium	0·0161	1·11	0·13	0·004	1·03	0·91	0·08
Lead	0·0034	1·15	0·06	0·0005	1·18	1·05	0·001
Tin	0·0021	1·24	0·04	0·0004	1·14	1·02	0·01
Sn+1% In	0·44	1·25	0·65	0·08	1·26	1·07	0·1
Sn+5·4% In	2·82	1·74	1·64	0·18	1·90	1·68	0·19
Brass	3·57	1·78	1·84	0·96	1·94	1·84	0·96
Eureka	44·9	6·32	6·33	5·62	6·33	6·33	5·62
Staybrite	47·8	8·23	6·53	5·66	6·53	6·53	5·66
Bi–Sn eutectic (C.S.)	7·08	7·63	2·57	1·20	2·69	2·55	1·20

M.P. = Mechanically polished with ‘ Brasso ’. C.S. = Cast surface.

The disagreement with the classical theory shown by the alloys Staybrite and the Bi–Sn eutectic is significant. Pure bismuth is known to have a high absorptivity (20%) at room temperature (theoretical value ~10%) and this suggests that the higher observed absorptivities of these alloys are due to phenomena such as the internal photoelectric effect. The agreement with theory shown by the other alloys, brass, Eureka and the 5·4% indium specimen, and the agreement of the absorptivity of Eureka with that (5·8%) deduced by Hagen and Rubens from reflectivity measurements at room temperature (the electrical resistivity of Eureka is practically independent of temperature over the range under consideration) seems to confirm the estimate of the effective intensity of the source.

Finally, the high value of the absorptivity of the 'Brasso'-polished copper, which is not very different from that (1·29%) obtained by Wexler (1951), emphasizes the importance of using electrolytically polished surfaces for such optical measurements. This is in agreement with the conclusion of Chambers that a layer of high resistivity is produced at the surface of a mechanically polished surface.

ACKNOWLEDGMENTS

I am indebted to Dr. D. Shoenberg and to Dr. A. B. Pippard for the suggestion of the problem and of many ideas during the course of this work. I am also grateful to the Royal Commission for the Exhibition of 1851 for the award of an Overseas Research Scholarship.

REFERENCES

Chambers, R. G., 1950 a, *Nature, Lond.*, **165,** 239; 1950 b, *Proc. Roy. Soc.* A, **202,** 378.
Daunt, J. G., Keeley, T. C., and Mendelssohn, K., 1937, *Phil. Mag.*, **23,** 264.
Kemsley, D. S., 1948, Physical Metallurgy Report 7, C.S.I.R., Melbourne.
London, H., 1940, *Proc. Roy. Soc.* A, **176,** 522.
Maxwell, E., Marcus, P. M., and Slater, J. C., 1949, Technical Report 109, Electronics Lab., M.I.T.
Pippard, A. B., 1947 a, *Proc. Roy. Soc.* A, **191,** 370; 1947 b, *Ibid.*, **191,** 385; 1949, *Physica*, **15,** 40; 1950, *Proc. Roy. Soc.* A, **203,** 98.
Reuter, G. E. H., and Sondheimer, E. H., 1948, *Proc. Roy. Soc.* A, **195,** 336.
Wexler, A., 1951, Scientific Paper 1558, Westinghouse Research Lab.

Antiferromagnetism by the Spin Wave Method
I: The Energy Levels

By J. M. ZIMAN
Clarendon Laboratory, Oxford

Communicated by M. H. L. Pryce; MS. received 4th February 1952

ABSTRACT. The Hamiltonian operator for an exchange antiferromagnet is expressed in terms of the deviations of the spins from the basic ordered antiparallel arrangement. By a Fourier transformation the Hamiltonian may be approximately represented as a sum of harmonic oscillator terms, corresponding to spin waves. The role of anisotropy energy in fixing the domain axis is discussed, and the energy levels are calculated for the case of an external field parallel to this axis. The ground-state energy includes the zero-point energy of the spin waves and is in good agreement with other theories. Formulae are quoted for a Hamiltonian including dipole–dipole interaction between non-isotropic spins.

§ 1. INTRODUCTION

One of the basic methods for the theoretical treatment of ferromagnetism is that of the 'spin wave', a concept due to Bloch (1930, 1932). Starting from the basic ordered state of the lattice, spin deviation operators are defined to describe the disordered states, and the Hamiltonian is expressed in terms of these. By a Fourier transformation of these deviation operators (and after making certain approximations) the Hamiltonian can be made to separate into a set of independent harmonic oscillator terms, each term corresponding to a wave of spin deviations propagated through the crystal by the exchange interaction between neighbours. In this way Bloch was able to deduce the temperature variation, and Holstein and Primakoff (1940) the field dependence, of the intrinsic magnetization.

The particular value of this method is that it is properly quantum-mechanical, and does not make the rather drastic assumption that the spins may be replaced by Ising dipoles. Moreover, it is not restricted to nearest-neighbour interactions—even long-range dipole–dipole interactions may be considered explicitly. The only limitation to its application is that the mean value of the spin deviation should be small; it is essentially a low-temperature theory.

The problem to be considered here is the possibility of extending this powerful technique to the case of antiferromagnetism, a term which is generally used to describe the phenomenon whereby certain substances tend to fall into ordered states in which the spins of neighbouring atoms are antiparallel (Van Vleck 1941, Grenoble Conference 1951). Such a state would seem natural for a substance in which exchange forces favour antiparallel spins on neighbouring or next-neighbouring sites; it has also been shown that for classical dipoles the antiparallel state may be the lowest, depending on the crystal structure and the shape of the specimen (Sauer 1940, Luttinger and Tisza 1946).

§2. THE ANTIFERROMAGNETIC LATTICE

The specimen is to consist of a crystalline lattice whose N sites are located by the values of the vector

$$\mathbf{R}_l = l_1\mathbf{u}_1 + l_2\mathbf{u}_2 + l_3\mathbf{u}_3, \qquad \ldots\ldots(1)$$

where l_1, l_2, l_3, are integers and $\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$ are the basic vectors of the lattice. On the lth site there is to be found a spin with operator $\mathbf{S}_l$ of total spin S (in units of $\hbar$). Between the lth and mth spins there is an exchange interaction $-2J_{lm}\mathbf{S}_l\cdot\mathbf{S}_m$ depending on the magnitude of the relative distance operator $\mathbf{R}_{lm} = \mathbf{R}_l - \mathbf{R}_m$. Thus the major term in the total Hamiltonian of the system is

$$\mathscr{H}_{\mathrm{E}} = -\tfrac{1}{2}\Sigma_{l,m}2J_{lm}\mathbf{S}_l\cdot\mathbf{S}_m. \qquad \ldots\ldots(2)$$

The problem of finding even the lowest eigenstate of $\mathscr{H}_{\mathrm{E}}$ is exceedingly difficult when J_{lm} is negative. But the lowest states for classical or Ising dipoles with the given Hamiltonian are known, and it is at least plausible that the eigenstate is derived from this by a small perturbation. In the ferromagnetic case the same assumption is made (quasi-saturation), although here also the completely saturated state is not an eigenstate of the complete Hamiltonian, owing to the magnetic interaction terms. The whole question has been considered in detail by Anderson (1951 a), using the Kramers–Heller spin-wave technique, and the assumption is thought to be justified.

To describe this basic state we must introduce a special vector $\mathbf{w}$ such that $\exp(i\mathbf{w}\cdot\mathbf{R}_l)$ alternates in sign from one lattice point to the next. An expression of this sort is sufficient to describe the antiparallel ordering due to strong nearest-neighbour interaction in the simple cubic and body-centred cubic lattices, but in other cases it may be necessary to take a linear combination of such terms (Luttinger 1951). This does not lead to any essential difficulty, but it will not be considered here because it introduces unnecessary complications.

The vector $\mathbf{w}$ is best defined in terms of the vectors reciprocal to $\mathbf{u}_1$, $\mathbf{u}_2$, $\mathbf{u}_3$, i.e. $\mathbf{w}=w_1\mathbf{v}_1+w_2\mathbf{v}_2+w_3\mathbf{v}_3$, where

$$\mathbf{v}_1\equiv\frac{\mathbf{u}_2\times\mathbf{u}_3}{\mathbf{u}_1\cdot\mathbf{u}_2\times\mathbf{u}_3},\quad \mathbf{v}_2\equiv\frac{\mathbf{u}_3\times\mathbf{u}_1}{\mathbf{u}_1\cdot\mathbf{u}_2\times\mathbf{u}_3},\quad \mathbf{v}_3\equiv\frac{\mathbf{u}_1\times\mathbf{u}_2}{\mathbf{u}_1\cdot\mathbf{u}_2\times\mathbf{u}_3}.$$

In this way, $\mathbf{R}_l\cdot\mathbf{w}=l_1w_1+l_2w_2+l_3w_3$, and in the typical case of the simple cubic lattice with next neighbour interactions $\mathbf{w}=\pi(\mathbf{i}+\mathbf{j}+\mathbf{k})$.

Besides this alternating function, we also need to prescribe a direction along which the spins would be aligned in order to fix the basic state exactly. It is usual to assume that a crystal of an antiferromagnet consists of domains, in each of which the spins are parallel or antiparallel to a given direction, this direction varying in a random manner from domain to domain. Yet the operator $\mathscr{H}_{\mathrm{E}}$ is isotropic, and there is no recognizable direction of alignment at any point of the crystal. When a field is applied externally, this provides a preferred direction, and the whole system will be affected equally, swinging round until it is in a position of minimum potential energy and destroying any assumed domain structure. In order to overcome this difficulty, it is necessary to assume that there is an additional anisotropy energy fixing certain directions of easy magnetization (Nagamiya 1951, Kittel 1951).

It is not easy to decide the origin of this term. If it is to be correctly described, with, for example, cubic symmetry in the cubic lattice, it would contain terms of the fourth degree in the spin operators and lead to very complicated expressions. The best that can be done for the present is to introduce into the Hamiltonian a term

$$\mathscr{H}_{\mathrm{A}}=-\Sigma_l g\beta S_l^{(z)}H_{\mathrm{a}}\exp(i\mathbf{w}\cdot\mathbf{R}_l), \qquad \ldots\ldots(3)$$

that is, a fictitious internal field staggered so as always to be parallel to the spin in the basic Ising state.* βH_{A} need only be very small compared with J_{lm} in order to make sure that the spin system as a whole maintains its direction. This direction, the z axis, will evidently be one of the crystallographic axes of the crystal. A more general condition for domain stability will be discussed later.

To calculate the parallel susceptibility we also need a magnetic field H_z (due to external sources) along the z axis, making the total Hamiltonian

$$\mathscr{H}=-\tfrac{1}{2}\Sigma_{l,m}2J_{lm}\mathbf{S}_l\cdot\mathbf{S}_m-\Sigma_l g\beta S_l^{(z)}\{H_z+H_{\mathrm{A}}\exp(i\mathbf{w}\cdot\mathbf{R}_l)\}. \qquad \ldots\ldots(4)$$

This expression does not include all possible types of interaction, but covers those which have been generally considered in the literature. The generalization to non-isotropic spins is discussed briefly in the Appendix.

§ 3. SPIN-DEVIATION REPRESENTATION

We shall use a representation in which the $S_l^{(z)}$ components are diagonal. The wave function for the whole lattice may then be considered as a linear combination of all the various product wave functions, one from each site, which can be labelled by the values of $S_l^{(z)}$. Let us for the moment consider only the sites for which the value of $S_l^{(z)}$ in the assumed basic state is positive, i.e. those which 'point to the right' in the lowest-energy configuration. We define the following operators:

$$S_l^{+}\equiv S_l^{(x)}+iS_l^{(y)},\quad S_l^{-}\equiv S_l^{(x)}-iS_l^{(y)},\quad \mathfrak{n}_l\equiv S-S_l^{(z)} \qquad \ldots\ldots(5)$$

---

* As usual, β is the Bohr magneton, and g the Landé factor for the spin.

$\mathfrak{n}_l$ is the spin deviation operator. The states, which were formerly labelled by values of $S_l^{(z)}$ may now be relabelled (on the right-hand sites, that is) by the eigenvalues n_l of $\mathfrak{n}_l$. Thus

$$\mathfrak{n}_l \,|\, S_1^{(z)} \ldots\ldots S_l^{(z)} \ldots\ldots S_N^{(z)} \rangle = \mathfrak{n}_l \,|\, n_1 \ldots\ldots n_l \ldots\ldots n_N \rangle$$

But
$$S_l^{+} \,|\, n_l \rangle = (2S)^{1/2} \left(1 - \frac{n_l - 1}{2S}\right)^{1/2} (n_l)^{1/2} \,|\, n_l - 1 \rangle$$

$$S_l^{-} \,|\, n_l \rangle = (2S)^{1/2} (n_l + 1)^{1/2} (1 - n_l/2S)^{1/2} \,|\, n_l + 1 \rangle \qquad \ldots\ldots(6)$$

and if we introduce creation and destruction operators a_l, a_l^*, where

$$a_l^* \,|\, n_e = (n_l + 1)^{1/2} \,|\, n_l + 1 \rangle, \quad a_l \,|\, n_l \rangle = (n_l)^{1/2} \,|\, n_l - 1 \rangle \qquad \ldots\ldots(7)$$

we find

$$\left.\begin{aligned} &\mathfrak{n}_l = a_l^* a_l \\ &S_l^{+} = (2S)^{1/2} (1 - a_l^* a_l/2S)^{1/2} a_l, \quad S_l^{-} = (2S)^{1/2} a_l^* (1 - a_l^* a_l/2S)^{1/2} \end{aligned}\right\} \ldots(8)$$

For the left-hand spins, i.e. those for which $\exp(i\mathbf{w} \,.\, \mathbf{R}_l) = -1$, we define our spin deviation operator in the opposite sense:

$$\mathfrak{n}_l' \equiv S + S_l^{(z)}. \qquad \ldots\ldots(9)$$

The states can be relabelled in terms of the eigenvalues n_l' of $\mathfrak{n}_l'$ and we have

$$\left.\begin{aligned} &\mathfrak{n}_l' \,|\, n_l' \rangle = n_l' \,|\, n_l' \rangle \\ &S_l^{+} \,|\, n_l' \rangle = (2S)^{1/2} (n_l' + 1)^{1/2} (1 - n_l'/2S)^{1/2} \,|\, n_l' + 1 \rangle \\ &S_l^{-} \,|\, n_l' \rangle = (2S)^{1/2} \{1 - (n_l' - 1)/2S\}^{1/2} \,|\, n_l' + 1 \rangle. \end{aligned}\right\} \ldots\ldots(10)$$

Creation and destruction operators are again defined by the equations

$$a_l^* \,|\, n_l' \rangle = (n_l' + 1)^{1/2} \,|\, n_l' + 1 \rangle, \quad a_l \,|\, n_l' \rangle = (n_l')^{1/2} \,|\, n_l' - 1 \rangle \qquad \ldots\ldots(11)$$

to give

$$\left.\begin{aligned} &\mathfrak{n}_l' = a_l^* a_l \\ &S_l^{+} = (2S)^{1/2} a_l^* (1 - a_l^* a_l/2S)^{1/2}, \quad S_l^{-} = (2S)^{1/2} (1 - a\,^* a_l/2S)^{1/2} a_l. \end{aligned}\right\} .(12)$$

This analysis parallels that of Holstein and Primakoff in the ferromagnetic case, and with them we shall assume that factors such as $(1 - a_l^* a_l/2S)^{1/2}$ may be dropped, since their expectation value is $(1 - n_l/2S)^{1/2}$ and the expectation value of the spin deviation is to be assumed to be very much smaller than its maximum $2S$. This is effectively assuming a low temperature, but the effect of zero-point motion must not be neglected, and the whole question is discussed in part II of the present paper (Ziman 1952). In the same way we shall neglect terms in the Hamiltonian of higher order than the second in a_l, a_l^*.

To this approximation eqns. (5), (8), (9) and (12) may be consolidated as follows:

$$\left.\begin{aligned} &S_l^{(x)} = (\tfrac{1}{2}S)^{1/2} (a_l + a_l^*) \\ &S_l^{(y)} = -i \exp(i\mathbf{w} \,.\, \mathbf{R}_l) (\tfrac{1}{2}S)^{1/2} (a_l - a_l^*) \\ &S_l^{(z)} = \exp(i\mathbf{w} \,.\, \mathbf{R}_l) (S - a_l^* a_l), \end{aligned}\right\} \qquad \ldots\ldots(13)$$

and putting these into the Hamiltonian (4) we have

$$\begin{aligned} \mathscr{H} = -\Sigma_{l,m} J_{lm} (\tfrac{1}{2}S) \{ &(a_l a_m + a_l^* a_m^* + a_l a_m^* + a_l^* a_m) \\ &- \sigma_{lm} (a_l a_m + a_l^* a_m^* - a_l a_m^* - a_l^* a_m + 2a_l^* a_l + 2a_m^* a_m) \} \\ &+ \Sigma_l g\beta \{ H_{\mathrm{A}} + H_z \exp(i\mathbf{w} \,.\, \mathbf{R}_l) \} a_l^* a_l + C \end{aligned} \qquad \ldots\ldots(14)$$

where
$$C = -\Sigma_{l,m} J_{lm} \sigma_{lm} S^2 - \Sigma_l g\beta S \{ H_{\mathrm{A}} + H \exp(i\mathbf{w} \,.\, \mathbf{R}_l) \} \qquad \ldots\ldots(15)$$

and
$$\sigma_{lm} = \exp(i\mathbf{w} \,.\, \mathbf{R}_{lm}).$$

.A
5
2

The term in C which depends on H_z will vanish; it represents the magnetic moment of the basic Ising array, which must be zero. We need no longer include C in the Hamiltonian, since it is merely a constant energy.

§ 4. SPIN-WAVE REPRESENTATION

In order to reduce the double summations in (14) we perform a Fourier transformation on the operators. Let

$$\left\{\begin{aligned} \alpha_\lambda &= N^{-1/2}\Sigma_l a_l \exp(i\mathbf{K}_\lambda . \mathbf{R}_l), \\ \alpha_\lambda{}^* &= N^{-1/2}\Sigma_l a_l{}^* \exp(-i\mathbf{K}_\lambda . \mathbf{R}_l), \end{aligned}\right. \text{ i.e. } \left\{\begin{aligned} a_l &= N^{-1/2}\Sigma_\lambda \alpha_\lambda \exp(-i\mathbf{K}_\lambda . \mathbf{R}_l), \\ a_l{}^* &= N^{-1/2}\Sigma_\lambda \alpha_\lambda{}^* \exp(i\mathbf{K}_\lambda . \mathbf{R}_l). \end{aligned}\right\} \quad \ldots(16)$$

$\mathbf{K}_\lambda$ is a wave vector, and the periodicity conditions require that it shall be of the form

$$\mathbf{K}_\lambda = 2\pi\left(\frac{\lambda_1}{G_1}\mathbf{v}_1 + \frac{\lambda_2}{G_2}\mathbf{v}_2 + \frac{\lambda_3}{G_3}\mathbf{v}_3\right) \quad \ldots\ldots(17)$$

where G_1, G_2, G_3 are the dimensions of the crystal measured in lattice spacings along the basis vectors and λ_1, λ_2, λ_3 are integers such that $-\tfrac{1}{2}G_1 \leqslant \lambda_1 \leqslant \tfrac{1}{2}G_1 - 1$, $-\tfrac{1}{2}G_2 \leqslant \lambda_2 \leqslant \tfrac{1}{2}G_2 - 1$, $-\tfrac{1}{2}G_3 \leqslant \lambda_3 \leqslant \tfrac{1}{2}G_3 - 1$.

To express the Hamiltonian in terms of these new operators we must evaluate terms such as

$$\sum_{l,m} f(\mathbf{R}_{lm})a_l{}^*a_m = N^{-1} \sum_{l,m,\lambda,\lambda'} f(\mathbf{R}_{lm})\alpha_{\lambda'}{}^*\alpha_\lambda \exp\{i(\mathbf{K}_\lambda . \mathbf{R}_l - \mathbf{K}_{\lambda'} . \mathbf{R}_m)\}.$$

Putting $\mathbf{R}_h = \mathbf{R}_{lm}$ we obtain

$$\sum_{l,h,\lambda,\lambda'} N^{-1}f(\mathbf{R}_h)\alpha_{\lambda'}{}^*\alpha_\lambda \exp\{i(\mathbf{K}_{\lambda'} - \mathbf{K}_\lambda) . \mathbf{R}_h\} \exp\{i\mathbf{K}_\lambda . \mathbf{R}_h\}.$$

Now the summations over l and h may be carried out independently so long as effects at the surface of the specimen may be neglected. In an antiferromagnet this is always allowable, since there is no intrinsic magnetization leading to shape-dependent effects. The summation over l merely gives $N\delta_{\lambda\lambda'}$, and the expression becomes

$$\Sigma_\lambda\{\Sigma_h f(\mathbf{R}_h) \exp(i\mathbf{K}_\lambda . \mathbf{R}_h)\}\alpha_\lambda{}^*\alpha_\lambda.$$

For terms containing the factor $\exp(i\mathbf{w} . \mathbf{R}_l)$ the result is somewhat different. Thus,

$$\begin{aligned} \sum_{l,m} f(\mathbf{R}_{lm})a_l{}^*a_m \exp(i\mathbf{w} . \mathbf{R}_l) &= \sum_{l,m,\lambda,\lambda'} N^{-1}f(\mathbf{R}_{lm})\alpha_{\lambda'}{}^*\alpha_\lambda \\ &\qquad \exp\{i(\mathbf{K}_{\lambda'} . \mathbf{R}_l - \mathbf{K}_\lambda . \mathbf{R}_m + \mathbf{w} . \mathbf{R}_l)\} \\ &= \sum_{l,h,\lambda,\lambda'} N^{-1}f(\mathbf{R}_h)\alpha_{\lambda'}{}^*\alpha_\lambda \\ &\qquad \exp\{i(\mathbf{K}_{\lambda'} - \mathbf{K}_\lambda + \mathbf{w}) . \mathbf{R}_l\} \exp(i\mathbf{K}_\lambda . \mathbf{R}_h) \\ &= \Sigma_\lambda\{\Sigma_h f(\mathbf{R}_h)\alpha_{\lambda'}{}^*\alpha_\lambda \exp(i\mathbf{K}_\lambda . \mathbf{R}_h)\} \end{aligned}$$

where λ' is obtained from λ by the equation

$$\mathbf{K}_{\lambda'} = \mathbf{K}_\lambda \pm \mathbf{w} \quad \ldots\ldots(18)$$

(the sign being chosen to bring $\mathbf{K}_{\lambda'}$ into the region defined for $\mathbf{K}_\lambda$).

Terms in a_la_m lead to terms in $\alpha_\lambda\alpha_{-\lambda}$, as can readily be verified, and by carrying through all the summations and transformations we finally obtain

$$\mathscr{H} = \Sigma_\lambda(A_\lambda\alpha_\lambda{}^*\alpha_\lambda + \tfrac{1}{2}B_\lambda\alpha_\lambda\alpha_{-\lambda} + \tfrac{1}{2}B_\lambda{}^*\alpha_\lambda{}^*\alpha_{-\lambda}{}^* + g\beta H_z\alpha_\lambda{}^*\alpha_{\lambda'}) \quad \ldots\ldots(19)$$

where

$$A_\lambda = g\beta H_A + \Sigma_h 2SJ_h\{\sigma_h - \tfrac{1}{2}(1 + \sigma_h)\cos(\mathbf{K}_\lambda . \mathbf{R}_h)\} \quad \ldots\ldots(20)$$

$$B_\lambda = -\Sigma_h SJ_h(1 - \sigma_h)\exp(i\mathbf{K}_\lambda . \mathbf{R}_h); \quad \ldots\ldots(21)$$

$B_\lambda{}^*$ is complex conjugate of B_λ; $J_h = J(\mathbf{R}_h)$; $\sigma_h = \sigma(\mathbf{R}_h) = \exp(i\mathbf{w} . \mathbf{R}_h)$.

In order to write this Hamiltonian without any concealed repetition of terms, the $\mathbf{K}_\lambda$ space is dissected into four distinct volumes, such that when $\mathbf{K}_\lambda$ lies in one, $\mathbf{K}_{\lambda'}$, $\mathbf{K}_{-\lambda}$, $\mathbf{K}_{-\lambda'}$ lie one in each of the others. Thus, in the standard example of the simple cubic lattice, we could take the regions defined by

$$0 \leqslant \mathbf{K}_\lambda^{(z)} < \tfrac{1}{2}\pi; \quad -\tfrac{1}{2}\pi \leqslant \mathbf{K}_{-\lambda}^{(z)} < 0; \quad -\pi \leqslant \mathbf{K}_{\lambda'}^{(z)} < -\tfrac{1}{2}\pi; \quad \tfrac{1}{2}\pi \leqslant \mathbf{K}_{-\lambda}^{(z)} < \pi.$$

The summation is now to be taken over the first region only, the operators being written with their appropriate labels to cover the other three regions. That is to say,

$$\begin{aligned}\mathscr{H} = \Sigma_\lambda\{&A_\lambda\alpha_\lambda{}^*\alpha_\lambda + A_{-\lambda}\alpha_{-\lambda}{}^*\alpha_{-\lambda} + A_{\lambda'}\alpha_{\lambda'}{}^*\alpha_{\lambda'} + A_{-\lambda'}\alpha_{-\lambda'}{}^*\alpha_{-\lambda'} \\ &+ B_\lambda\alpha_\lambda\alpha_{-\lambda} + B_\lambda{}^*\alpha_\lambda{}^*\alpha_{-\lambda}{}^* + B_{\lambda'}\alpha_{\lambda'}\alpha_{-\lambda'} + B_{\lambda'}{}^*\alpha_{\lambda'}{}^*\alpha_{-\lambda'}{}^* \\ &+ g\beta H_z(\alpha_\lambda{}^*\alpha_{\lambda'} + \alpha_{-\lambda}{}^*\alpha_{-\lambda'} + \alpha_{\lambda'}{}^*\alpha_\lambda + \alpha_{-\lambda'}{}^*\alpha_{-\lambda})\}. \qquad \ldots\ldots(22)\end{aligned}$$

This expression contains no double sums, and no cross-products except those involving four operators all belonging to a definite value of λ. Each such set may be diagonalized by a procedure due to Polder (1949), which leaves the commutation properties unaltered. We find that

$$\mathscr{H} = \Sigma_\lambda\{u_\lambda{}^+(C_+{}^*(\lambda)C_+(\lambda) + \tfrac{1}{2}) + \mu_\lambda{}^-(C^*_-(\lambda)C_-(\lambda) + \tfrac{1}{2}) - (A_\lambda + A_{\lambda'})\} \quad \ldots\ldots(23)$$

where
$$\mu_\lambda^\pm = (A_\lambda{}^2 - |B_\lambda|^2)^{1/2} \pm g\beta H_z \qquad \ldots\ldots(24)$$

and
$$C_\pm{}^*(\lambda)C_\pm(\rho) - C_\pm(\rho)C_\pm{}^*(\lambda) = \delta_{\lambda\rho}. \qquad \ldots\ldots(25)$$

The summation is here over the half space $0 \leqslant \mathbf{K}_\lambda^{(z)} < \pi$.

These equations have a simple physical significance. The operator $C_\pm{}^*(\lambda)C_\pm(\lambda)$ has positive integral eigenvalues, $N_\lambda{}^\pm = 0, 1, 2, \ldots$. The corresponding eigenfunctions are standing waves of spin deviations, with wave vector $\mathbf{K}_\lambda$, propagated throughout the lattice; each spin wave quantum contributes an energy $\mu_\lambda{}^\pm$. The two signs in (24) show that the spin waves fall into pairs, whose energy levels are altered in opposite directions by the external magnetic field, according to whether the spin deviations they cause are aided or opposed by this field.

In this spin-wave representation the role of the anisotropy field is clear. If both H_z and H_A were zero, reference to (20) and (21) would show that for certain values of λ (in particular for $\mathbf{K}_\lambda = 0, \pm\mathbf{w}$) the spin waves have zero energy. This would mean that any number of 'quanta' could be put into those modes, turning the system out of the z direction, making the approximations invalid and the quantization in a particular direction meaningless. The significance of H_A is that it prevents $\mu_\lambda{}^\pm$ from vanishing for any λ, and we may assert this as a general requirement rather than proposing a definite form for the anisotropy. We should also note that when H_z is sufficiently large we may find that $\mu_\lambda{}^-$ vanishes. This would correspond to the tendency for a domain to set itself perpendicular to the applied field, as pointed out by Hulthén (1936). But the field necessary for this is much larger than H_A, since $(A_\lambda{}^2 - |B_\lambda|^2)^{1/2}$ is at least of the order $(g\beta H_A J_h)^{1/2}$ and $J_h \gg \beta H_A$.

§ 5. THE GROUND STATE

We have shown that the energy levels of our system may be represented by the formula
$$E = \Sigma_\lambda\{(N_\lambda{}^+ + \tfrac{1}{2})\mu_\lambda{}^+ + (N_\lambda{}^- + \tfrac{1}{2})\mu_\lambda{}^- - (A_\lambda + A_{\lambda'})\}. \qquad \ldots\ldots(26)$$

It has been pointed out that alternating Ising model basic state with which we started is not an eigenstate of the Hamiltonian. But if, in (26), we were to take each $N_\lambda^\pm$ to be zero, we should obtain the lowest energy level of the system, and the corresponding eigenstate would be our estimate of the lowest, or ground state. This differs from the basic state by the zero-point motion corresponding to a half quantum in each spin-wave mode. It would be possible to represent this state in terms of spin deviations, but the result would not have very great significance; the spin-wave representation is the more natural description.

What is of particular interest is the actual value of the energy of the ground state. Using the parallel ordered state (energy $-N\Sigma_h J_h S^2$) as a reference level, and remembering the term C dropped from (14), we have (in zero external field)

$$E_0 = N\Sigma_h J_h S^2(1-\sigma_h) + \tfrac{1}{2}\Sigma_\lambda\{(\mu_\lambda^+ + \mu_\lambda^-) - 2(A_\lambda + A_{\lambda'})\}. \qquad \ldots\ldots(27)$$

This may be evaluated by replacing the summation by an integral in any particular case. It has been shown by Anderson (1951 b) that for a system with nearest neighbour interactions J between two distinct sub-lattices

$$E_0 = 2ZJNS^2(1+\gamma/2ZS) \qquad \ldots\ldots(28)$$

where Z is the number of nearest neighbours and γ lies between 0 and 1. From (27) we have calculated the values of γ shown in the table.

Lattice type	Z	γ
Linear chain	2	0·726
Square plane	4	0·612
Simple cubic	6	0·57
Body-centred cubic	8	0·59

(The anisotropy has been neglected in this calculation.)

These results agree entirely with those obtained by Anderson (1951 a) using the Kramers–Heller technique. It is encouraging to note that they lie near the centre of the range of variation allowed by (28) and that the estimated ground state energy for the linear chain is close to the value corresponding to $\gamma = 0{\cdot}7726$ found by Hulthén (1938) by an exact calculation for spin ½. The difference between E_0 and the Ising value is quite small for large co-ordination number; for the simple cubic lattice it is about 9·5%. This suggests that the basic state is a good starting point for an approximate method, in agreement with the neutron diffraction experiments (Shull, Strauser and Wollan 1951) which have demonstrated the reality of the alternating lattice picture.

APPENDIX

These calculations may be generalized to include more complex forms of interaction. Thus, in place of (4), we may take the following Hamiltonian:

$$\begin{aligned}\mathscr{H} = &-\tfrac{1}{2}\Sigma_{l,m}2J_{lm}\mathbf{S}_l\,.\,\mathbf{S}_m - \Sigma_l g_z\beta S_l^{(z)}\{H_z + H_A \exp(i\mathbf{w}\,.\,\mathbf{R}_l)\}\\ &+\tfrac{1}{2}\Sigma_{l,m}\beta^2\mathbf{R}_{lm}^{-5}\{\mathbf{R}_{lm}^2\mathbf{S}_l\,.\,\mathbf{G}^2\,.\,\mathbf{S}_m - 3\mathbf{S}_l\,.\,\mathbf{G}\,.\,\mathbf{R}_{lm}\mathbf{S}_m\,.\,\mathbf{G}\,.\,\mathbf{R}_{lm}\} \qquad \ldots\ldots(29)\end{aligned}$$

where $\mathbf{G}$ is the tensor of the Landé g factors for non-isotropic spins. This is the most general form for the inclusion of dipole–dipole interactions. In order to make certain sums vanish, we must insist that $\mathbf{G}$ shall have its principal axes along the crystallographic axes x, y, z, but this is not a very stringent restriction. Putting

$$\mathbf{G} = g_x\mathbf{ii} + g_y\mathbf{jj} + g_z\mathbf{kk} \qquad \ldots\ldots(30)$$

and making the same substitutions and transformations as before, we get,

$$\begin{aligned}\mathscr{H}=\Sigma_\lambda(A_\lambda\alpha_\lambda{}^*\alpha_\lambda+\tfrac{1}{2}B_\lambda\alpha_\lambda\alpha_{-\lambda}+\tfrac{1}{2}B_\lambda{}^*\alpha_\lambda{}^*\alpha_{-\lambda}{}^*+C_\lambda\alpha_\lambda{}^*\alpha_{\lambda'}+\tfrac{1}{2}D_\lambda\alpha_\lambda\alpha_{-\lambda'}\\+\tfrac{1}{2}D_\lambda{}^*\alpha_\lambda{}^*\alpha_{-\lambda'}{}^*)\end{aligned} \qquad \ldots\ldots(31)$$

where

$$\begin{aligned}A_\lambda=g_z\beta H_A+\Sigma_h[\{2SJ_h-S\beta^2R_h{}^{-5}g_z{}^2(R_h{}^2-3Z_h{}^2)\}\sigma_h-SJ_h(1+\sigma_h)\cos(\mathbf{K}_\lambda\,.\,\mathbf{R}_h)\\+\tfrac{1}{2}S\beta^2R_h{}^{-5}\{g_x{}^2(R_h{}^2-3X_h{}^2)+g_y{}^2\sigma_h(R_h{}^2-3Y_h{}^2)\}\cos(\mathbf{K}_\lambda\,.\,\mathbf{R}_h)]\end{aligned} \quad .(32)$$

$$\begin{aligned}B_\lambda=\Sigma_h[-SJ_h(1-\sigma_h)+\tfrac{1}{2}S\beta^2R_h{}^{-5}\{g_x{}^2(R_h{}^2-3X_h{}^2)-g_y{}^2\sigma_h(R_h{}^2-3Y_h{}^2)\}]\\\times\exp(i\mathbf{K}_\lambda\,.\,\mathbf{R}_h)\end{aligned} \qquad \ldots\ldots(33)$$

$$C_\lambda=g_z\beta H_z+i\Sigma_h\tfrac{3}{2}S\beta^2R_h{}^{-5}g_xg_y(1-\sigma_h)X_hY_h\exp(i\mathbf{K}_\lambda\,.\,\mathbf{R}_h) \qquad \ldots\ldots(34)$$

$$D_\lambda=i\Sigma_h\tfrac{3}{2}S\beta^2R_h{}^{-5}g_xg_y(1+\sigma_h)X_hY_h\exp(i\mathbf{K}_\lambda\,.\,\mathbf{R}_h) \qquad \ldots\ldots(35)$$

(X_h, Y_h, Z_h are the components of R_h along the crystallographic axes—not necessarily the generating vectors of the lattice).

The same type of reduction process leads to an equation identical with (23), except that $\mu_\lambda{}^\pm$ is now defined by

$$\begin{aligned}(\mu_\lambda{}^\pm)^2=\tfrac{1}{2}[(A_\lambda{}^2-|B_\lambda|^2)+(A_{\lambda'}|^2-|B_{\lambda'}|^2)+2|C_\lambda|^2-2D_\lambda{}^2\\\pm\{(A_\lambda{}^2-|B_\lambda|^2-A_{\lambda'}{}^2+|B_{\lambda'}|^2)^2+4E_\lambda{}^2([A_\lambda+A_{\lambda'}]^2-[B_\lambda-B_{\lambda'}]^2)^2\\+4(F_\lambda[A_\lambda+A_{\lambda'}-B_\lambda+B_{\lambda'}]-iD_\lambda[A_\lambda-A_{\lambda'}-B_\lambda+B_{\lambda'}])\\\times(F_\lambda[A_\lambda+A_{\lambda'}+B_\lambda+B_{\lambda'}]+iD_\lambda[A_\lambda-A_{\lambda'}+B_\lambda-B_{\lambda'}])\}^{1/2}]\end{aligned} \qquad \ldots\ldots(36)$$

where $C_\lambda=E_\lambda+iF_\lambda$.

It is not obvious from these equations whether $\mu_\lambda{}^\pm$ will always vanish for some values of λ when $H_A=0$, and it may be possible to interpret the anisotropy energy in terms of the dipole–dipole interaction, but this is a topic which will not be pursued here.

REFERENCES

ANDERSON, P. W., 1951 a, *An Approximate Quantum Theory of the Antiferromagnetic Ground State* (Bell Telephone Laboratories Report); 1951 b, *Phys. Rev.*, **83**, 1260 (L).

BLOCH, F., 1930, *Z. Phys.*, **61**, 206; 1932, *Ibid.*, **74**, 295.

GRENOBLE CONFERENCE, 1951, *J. Phys. Radium*, **12**, 149.

HOLSTEIN, T., and PRIMAKOFF, H., 1940, *Phys. Rev.*, **58**, 1098.

HULTHÉN, L., 1936, *Proc. Amst. Acad. Sci.*, **39**, 190; 1938, *Ark. Mat. Astr. Fys.*, **26**A, No. 11.

KITTEL, C., 1951, *Phys. Rev.*, **82**, 565 (L).

LUTTINGER, J. M., 1951, *Phys. Rev.*, **81**, 1015.

LUTTINGER, J. M., and TISZA, L., 1946, *Phys. Rev.*, **70**, 954.

NAGAMIYA, T., 1951, *Prog. Theor. Phys.*, **6**, 342.

POLDER, D., 1949, *Phil. Mag.*, **40**, 99.

SAUER, J. A., 1940, *Phys. Rev.*, **57**, 142.

SHULL, C. G., STRAUSER, W. A., and WOLLAN, E. O., 1951, *Phys. Rev.*, **83**, 333.

VAN VLECK, J. H., 1941, *J. Chem. Phys.*, **9**, 85.

ZIMAN, J. M., 1952, *Proc. Phys. Soc.* A, **65**, 548.

Antiferromagnetism by the Spin Wave Method
II: Magnetic Properties

By J. M. ZIMAN
Clarendon Laboratory, Oxford

Communicated by M. H. L. Pryce; MS. received 4th February 1952

ABSTRACT. From the energy levels obtained in a previous paper by the author the partition function and magnetic susceptibilities are calculated. $\chi_{\parallel}$ behaves like aT^2 at low temperatures, whilst $\chi_{\perp}$ is constant. The magnetic resonance absorption frequencies are also calculated and shown to depend strongly on the anisotropy field. By representing the true Hamiltonian in terms of the operators of the approximate Hamiltonian, correction is made for the disordering of the system by the spin waves, and susceptibility curves agreeing well with experiment can be calculated. The method is applicable to different types of lattice and of interaction, is not confined to zero fields, and is valid up to near the critical temperature.

---

§ 1. THE PARALLEL SUSCEPTIBILITY

In the previous paper of this series (Ziman 1952, to be referred to as I) we have shown that the energy levels for an antiferromagnet near the ground state may be expressed approximately in the form

$$E=\Sigma_\lambda\{(N_\lambda^{+}+\tfrac{1}{2})\mu_\lambda^{+}+(N_\lambda^{-}+\tfrac{1}{2})\mu_\lambda^{-}\}, \qquad \ldots\ldots(1)$$

where

$$\mu_\lambda^{\pm}=(A_\lambda^2-|B_\lambda|^2)^{1/2}\pm g\beta H_z \qquad \ldots\ldots(2)$$

and the $N_\lambda^{\pm}$ are integers equal to or greater than 0. (Constant terms have been dropped.)

The partition function may thus be written:

$$\begin{aligned}(\text{P.F.})&=\sum_{N_\lambda^{+},\,N_\lambda^{-}}\exp(-E/\boldsymbol{k}T)\\ &=\sum_{N_\lambda^{+},\,N_\lambda^{-}}[\exp\{-\Sigma_\lambda(N_\lambda^{+}+\tfrac{1}{2})\mu_\lambda^{+}/\boldsymbol{k}T\}\exp\{-\Sigma_\lambda(N_\lambda^{-}+\tfrac{1}{2})\mu_\lambda^{-}/\boldsymbol{k}T\}]\\ &=\Pi_\lambda\{4\ \operatorname{cosech}(\mu_\lambda^{+}/2\boldsymbol{k}T)\operatorname{cosech}(\mu_\lambda^{-}/2\boldsymbol{k}T)\}. \qquad \ldots\ldots(3)\end{aligned}$$

Hence the magnetization per unit volume is given by

$$M=\frac{\boldsymbol{k}T}{V}\frac{\partial}{\partial H_z}\ln(\text{P.F.})=-\frac{g\beta}{2V}\Sigma_\lambda\{\coth(\mu_\lambda^{+}/2\boldsymbol{k}T)-\coth(\mu_\lambda^{-}/2\boldsymbol{k}T)\}, \qquad \ldots\ldots(4)$$

where V is the volume of the crystal, and $\chi_{\parallel}$, the differential susceptibility, is obtained from

$$\chi_{\parallel}=\frac{\partial M}{\partial H_z}=\frac{g^2\beta^2}{4V\boldsymbol{k}T}\Sigma_\lambda\{\operatorname{cosech}^2(\mu_\lambda^{+}/2\boldsymbol{k}T)+\operatorname{cosech}^2(\mu_\lambda^{-}/2\boldsymbol{k}T)\}. \qquad \ldots\ldots(5)$$

In the limit, when the crystal is large, we may replace the sum over λ by an integral through the volume spanned by the wave vector. Thus, since $\mathbf{K}_\lambda=2\pi\{(\lambda_1/G_1)\mathbf{v}_1+(\lambda_2/G_2)\mathbf{v}_2+(\lambda_3/G_3)\mathbf{v}_3\}$, where λ_1, λ_2, λ_3, are integers, we have

$$\chi_{\parallel}=\frac{N}{V}\frac{g^2\beta^2}{\boldsymbol{k}T}\frac{1}{64\pi^3}\iiint_{-\pi}^{\pi}\{\operatorname{cosech}^2(\mu_\lambda^{+}/2\boldsymbol{k}T)+\operatorname{cosech}^2(\mu_\lambda^{-}/2\boldsymbol{k}T)\}\,d\mathbf{K}_\lambda, \qquad \ldots\ldots(6)$$

where $N = G_1 G_2 G_3$ and we have extended the integration over the whole space of $\mathbf{K}_\lambda$ and divided by 2, since the summation was over only half this volume.

This formula is quite general. It represents what is usually known as the parallel susceptibility—the susceptibility for a magnetic field applied parallel to the domain axis. The only difficulty is to evaluate the triple integral; in simple cases this can be done numerically. In the absence of anisotropy energy, or in very strong magnetic fields, the integrand will have singularities where $\mu_\lambda^\pm = 0$ and the integral may not converge. This is a convenient criterion for the stability of the system. Thus, if $\chi_\parallel$ does not converge even in a vanishingly small field, it suggests that the system is not stable against thermal agitation, and antiferromagnetism will not appear. For example, it is easy to see that the linear chain, and any plane lattice with only short-range interaction, should not be antiferromagnetic (cf. Yin-Yuan Li 1951).

.A
5
2

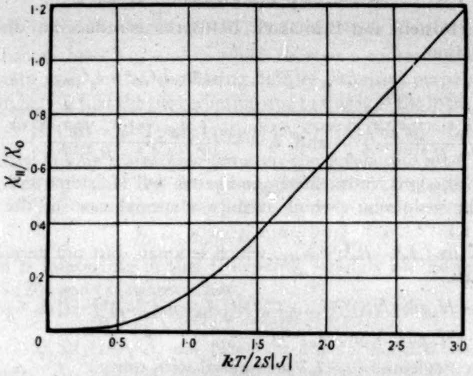


Fig. 1. Parallel susceptibility (uncorrected) for simple cubic lattice.

For the simple cubic lattice, with nearest neighbour interaction J and negligible anisotropy field, we have

$$\mu_\lambda^\pm = 4S|J|\{9-(\cos K_\lambda^{(1)} + \cos K_\lambda^{(2)} + \cos K_\lambda^{(3)})^2\}^{1/2} \pm g\beta H_z \quad \ldots\ldots(7)$$

and $\chi_\parallel$ converges if $H_z = 0$. Such a lattice is thus spontaneously antiferromagnetic; for fig. 1 we have computed $\chi_\parallel/\chi_0$ as a function of $\boldsymbol{k}T/2S|J|$, where

$$\chi_0 = Ng^2\beta^2/24VS|J|. \quad \ldots\ldots(8)$$

At low temperatures $\chi_\parallel$ behaves like $(\boldsymbol{k}T/2S|J|)^2\chi_0/6\sqrt{3}$, but becomes approximately linear in T as the temperature increases. No singularity corresponding to a Curie point appears; this is not surprising since the analysis is only valid at low temperatures. If we had included a small anisotropy term in this calculation the susceptibility would be only slightly reduced; its effect is only appreciable near poles of the integrand, and these contribute important terms only at very low temperatures.

§ 2. THE PERPENDICULAR SUSCEPTIBILITY

We now consider the effect of an external field H_x in the x direction, i.e. perpendicular to the domain axis. This contributes a term

$$\mathscr{H}' = -H_x g\beta \Sigma_l S_l^{(x)} \qquad \ldots\ldots(9)$$

to the Hamiltonian, and we shall treat this as a perturbation on our main Hamiltonian (for simplicity, in the absence of H_z). From eqns. (13) and (16) in I we have

$$\begin{aligned}\mathscr{H}' &= -H_x g\beta(\tfrac{1}{2}S)^{1/2}\Sigma_l(a_l + a_l^*)\\ &= -H_x g\beta(\tfrac{1}{2}S/N)^{1/2}\Sigma_{\lambda l}\{\exp(-i\mathbf{K}_\lambda . \mathbf{R}_l)\alpha_\lambda + \exp(i\mathbf{K}_\lambda . \mathbf{R}_l)\alpha_\lambda^*\}\\ &= -H_x g\beta(\tfrac{1}{2}NS)^{1/2}(\alpha_0 + \alpha_0^*) \qquad \ldots\ldots(10)\end{aligned}$$

where α_0 is the operator α_λ for $\mathbf{K}_\lambda = 0$.

This commutes with the whole of $\mathscr{H}$ except

$$\mathscr{H}_0 = A_0\alpha_0^*\alpha_0 + \tfrac{1}{2}B_0(\alpha_0\alpha_0 + \alpha_0^*\alpha_0^*). \qquad \ldots\ldots(11)$$

As shown by Holstein and Primakoff (1940), this is reduced to diagonal form by the transformation

$$\alpha_0 = l_1 C_0 + l_2 C_0^*, \quad \alpha_0^* = l_1 C_0^* + l_2 C_0,$$

where

$$l_1 = \left\{\frac{1}{2}\frac{A_0 + (A_0^2 - B_0^2)^{1/2}}{(A_0^2 - B_0^2)^{1/2}}\right\}^{1/2} \text{ and } l_2 = -\left\{\frac{1}{2}\frac{A_0 - (A_0^2 - B_0^2)^{1/2}}{(A_0^2 - B_0^2)^{1/2}}\right\}^{1/2}. \qquad \ldots\ldots(12)$$

(This choice of signs, not clearly indicated by Holstein and Primakoff, is essential to the reduction—which is only a special case of the method used previously in I.)

We may write $(A_0^2 - B_0^2)^{1/2} = \mu_0$, which is small, but not zero if our domain is stable. Thus

$$\begin{aligned}\mathscr{H}' &= -H_x g\beta(\tfrac{1}{2}NS)^{1/2}(C_0 + C_0^*)\{([A_0 + \mu_0]/2\mu_0)^{1/2} - ([A_0 - \mu_0]/2\mu_0)^{1/2}\}\\ &\simeq -H_x g\beta\tfrac{1}{2}(NS\mu_0/A_0)^{1/2}(C_0 + C_0^*) \qquad \ldots\ldots(13)\end{aligned}$$

where we have neglected $(\mu_0/A_0)^2$ compared with unity.

The part $\mathscr{H}_0$ of the Hamiltonian now becomes

$$\mathscr{H}_0 = \mu_0(C_0^*C_0 + \tfrac{1}{2}). \qquad \ldots\ldots(14)$$

If the eigenfunctions of $C_0^*C_0$ are $|N_0\rangle$, then

$$C_0|N_0\rangle = (N_0)^{1/2}|N_0 - 1\rangle, \; C_0^*|N_0\rangle = (N_0 + 1)^{1/2}|N_0 + 1\rangle$$

and clearly $\langle N_0|\mathscr{H}'|N_0\rangle = 0$ for all N_0.

But the second-order perturbation does not vanish, and the state $|N_0\rangle$ is shifted in energy by the amount

$$\begin{aligned}\Delta E &= \sum_{N_0' \neq N_0} \frac{\langle N_0|\mathscr{H}'|N_0'\rangle\langle N_0'|\mathscr{H}'|N_0\rangle}{\mu_0(N_0 - N_0')}\\ &= \frac{H_x^2 g^2\beta^2 NS\mu_0}{4A_0\mu_0} \sum_{N_0' \neq N_0} \frac{\langle N_0|C_0 + C_0^*|N_0'\rangle\langle N_0'|C_0 + C_0^*|N_0\rangle}{N_0 - N_0'} \quad ..(15)\\ &= -NSg^2\beta^2 H_x^2/4A_0.\end{aligned}$$

Thus the application of a perpendicular field alters the energy of the system by ΔE, regardless of the state of occupation of the levels, and we have

$$\chi_\perp = -\frac{1}{V}\frac{\partial^2}{\partial H_x^2}(\Delta E) = \frac{N}{V}\frac{g^2\beta^2 S}{2A_0}. \qquad \ldots\ldots(16)$$

In the case of the simple cubic lattice

$$\chi_\perp = S\chi_0. \qquad \ldots\ldots(17)$$

Hulthén (1936), using the Kramers–Heller technique, found

$$\chi_\perp = S\chi_0\{1-(\boldsymbol{k}T/2SJ)^2/3\sqrt{3}\}, \qquad \ldots\ldots(18)$$

which differs from (17) by the temperature-dependent term. The discrepancy presumably lies in the fact that the method used by Hulthén is a quantization of a semi-classical model, and his operators ξ and η are harmonic conjugates only when the system is unperturbed; when the state is partially disordered by temperature agitation their commutation properties do not remain constant. It will be shown later that corrections for the approximations in our Hamiltonian do not alter (16). It is interesting to observe that the anisotropy energy does not appear in (16)—or only as a small correction in (13). Yet it must always be included as a convergence factor, even if negligibly small.

§3. MAGNETIC RESONANCE

Suppose that we have a crystal in which there is a constant magnetic field H_z along the domain axis, and an oscillating field $H_x \exp(i\nu t)$ perpendicular to this. We have shown in (10) that the perturbation due to this is of the form

$$\mathscr{H}' = -H_x \exp(i\nu t) g\beta(\tfrac{1}{2}NS)^{1/2}(\alpha_0+\alpha_0^*), \qquad \ldots\ldots(19)$$

and this can only induce transitions between the levels due to $\mathscr{H}_0 + g\beta H_z\alpha_0^*\alpha_0'$, where $\alpha_0' = \alpha_{\chi'}$ for $\mathbf{K}_{\chi'} = \mathbf{w}$. But the spacing of these levels is uniform, and given by $\mu_0 \pm g\beta H_z$. Thus resonance absorption can only take place at frequencies

$$\boldsymbol{h}\nu = (A_0^2 - B_0^2)^{1/2} \pm g\beta H_z. \qquad \ldots\ldots(20)$$

This result is interesting in that it depends strongly on the anisotropy field. Reference to I (20) and (21) shows that

$$\boldsymbol{h}\nu = g\beta\sqrt{\{(H_A + 2H_E)H_A\}} \pm g\beta H_z, \qquad \ldots\ldots(21)$$

where $g\beta H_E = -\Sigma_h SJ_h(1-\sigma_h)$, and plays the role of an effective internal field due to exchange. An identical result has been obtained by Kittel (1951), using a sub-lattice internal field model, and it should be capable of experimental verification.

If the constant field does not happen to be parallel to the domain axis, only its component in that direction is effective in splitting the levels, whilst only the component of the alternating field in the xy plane is capable of inducing transitions. Experiments on powders, or on crystals in which the domain axes are randomly oriented, will thus show very broad lines; the two components of (21) may not be distinct (Nagamiya 1951).

§4. CORRECTION FOR SPIN-WAVE SATURATION

Up to now certain terms in the Hamiltonian have been ignored. These are terms introduced by factors such as $(1-a_l^*a_l/2S)^{1/2}$, which were dropped from I (13) and (14). So long as the average spin deviation is small compared with $2S$ this is perhaps permissible, but we must now try to correct for the approximation.

The neglected factor has a simple physical significance; it represents the fact that a given spin cannot acquire more than $2S$ deviations; it corresponds to interaction between the spin waves, which have hitherto been considered independent. We have assumed that any number of spin waves may be put into the system, whereas, in fact, beyond a certain point the lattice becomes saturated with them, and an additional quantum does not so much increase the energy but redistribute it from point to point. In the summations over $N_\lambda^{\pm}$ in the partition function we have allowed an infinity of values, where in fact we know that there cannot be more than N^{2S+1} states for the system as a whole.

Referring to eqns. (4), (5), (8) and (12) of I, we can see that the complete Hamiltonian should be written

$$\begin{aligned}\mathscr{H}^* = -\Sigma_{lm} J_{lm} \tfrac{1}{2} S \Bigg[&(1-\sigma_{lm}) \left\{ \left(1 - \frac{a_l^* a_l}{2S}\right)^{1/2} \left(1 - \frac{a_m^* a_m}{2S}\right)^{1/2} a_l a_m \right. \\ &\left. + a_l a_m^* \left(1 - \frac{a_l^* a_l}{2S}\right)^{1/2} \left(1 - \frac{a_m^* a_m}{2S}\right)^{1/2} \right\} + (1+\sigma_{lm}) \left\{ \left(1 - \frac{a_l^* a_l}{2S}\right)^{1/2} a_l a_m^* \right. \\ &\left. \times \left(1 - \frac{a_m^* a_m}{2S}\right)^{1/2} + a_l^* \left(1 - \frac{a_l^* a_l}{2S}\right)^{1/2} \left(1 - \frac{a_m^* a_m}{2S}\right)^{1/2} a_m \right\} \\ &- \sigma_{lm} S \left\{ a_l^* a_l \left(1 - \frac{a_m^* a_m}{2S}\right) + a_m^* a_m \left(1 - \frac{a_l^* a_l}{2S}\right) \right\} \Bigg] \\ &+ \Sigma_l g\beta \{H_A + H_s \sigma_l\} a_l^* a_l + C. \qquad \ldots\ldots(22)\end{aligned}$$

We now propose to replace $1 - a_l^* a_l/2S$ by $\phi = 1 - \langle a_l^* a_l \rangle_{\text{av}}/2S$ where $\langle a_l^* a_l \rangle_{\text{av}}$ is the expectation value of $a_l^* a_l$, averaged over all the sites, in a given spin-wave state. We ignore the non-commutation of a_l and $(1 - a_l^* a_l/2S)^{1/2}$, etc., and also the correlation factor which makes $\langle a_l^* a_l a_m^* a_m \rangle_{\text{av}}$ differ from $\langle a_l^* a_l \rangle_{\text{av}} \langle a_m^* a_m \rangle_{\text{av}}$ when l and m are close together. (This is justified in the Appendix.) Clearly

$$\mathscr{H}^* \simeq \phi H_E + \Sigma_l g\beta \{H_A + H_s \exp(i\mathbf{w} \cdot \mathbf{R}_l)\} a_l^* a_l, \qquad \ldots\ldots(23)$$

where ϕ is a number which depends on the state of occupation of the eigenstates $|N_\lambda^{\pm}\rangle$ of the approximate Hamiltonian $\mathscr{H}$.

We calculate ϕ as follows. The average of $a_l^* a_l$ over all sites is defined by

$$\begin{aligned}(a_l^* a_l)_{\text{av}} &= N^{-1} \Sigma_l a_l^* a_l \\ &= N^{-2} \sum_{l,\lambda,\lambda'} \exp\{i(\mathbf{K}_\lambda - \mathbf{K}_{\lambda'}) \cdot \mathbf{R}_l\} \alpha_\lambda^* \alpha_{\lambda'} \\ &= N^{-1} \Sigma_\lambda \alpha_\lambda^* \alpha_\lambda. \qquad \ldots\ldots(24)\end{aligned}$$

To find the expectation value of this operator we must transform to a representation in terms of $C_{\pm}(\lambda)$. In the general case, when H_s does not vanish, this may be rather complicated, but it is sufficient to ignore this term and consider the system in the absence of a field. The transformation is then of the form used by Holstein and Primakoff (e.g. eqn. (12)), and we have

$$\begin{aligned}(a_l^* a_l)_{\text{av}} = N^{-1} \Sigma_\lambda \{ &l_1^2 (C_\lambda^* C_\lambda + C_{-\lambda}^* C_{-\lambda}) + l_2^2 (C_\lambda C_\lambda^* + C_{-\lambda} C_{-\lambda}^*) \\ &+ l_1 l_2 (C_\lambda^* C_\lambda^* + C_\lambda C_\lambda - C_{-\lambda}^* C_{-\lambda}^* - C_{-\lambda} C_{-\lambda}) \}, \qquad \ldots\ldots(25)\end{aligned}$$

where the summation is over the half space of $\mathbf{K}_\lambda$ and l_1 and l_2 are defined as in (12), but with A_λ in place of A_0, etc.

The terms in $C_\lambda^* C_\lambda^*$, etc., do not contribute to the expectation value because $\langle N_\lambda | C_\lambda^* C_\lambda^* | N_\lambda \rangle = 0$, and we have

$$\begin{aligned}\langle N_\lambda | (a_l^* a_l)_{\text{av}} | N_\lambda \rangle &= N^{-1} \Sigma_\lambda \langle N_\lambda | l_1^2 C_\lambda^* C_\lambda + l_2^2 C_\lambda C_\lambda^* | N_\lambda \rangle \\ &= N^{-1} \Sigma_\lambda \{ (N_\lambda + \tfrac{1}{2}) A_\lambda/\mu_\lambda - \tfrac{1}{2} \} \qquad \ldots\ldots(26)\end{aligned}$$

where now the sum is over all λ, and $\mu_\lambda = (A_\lambda^2 - |B_\lambda|^2)^{1/2}$.

We have seen that the anisotropy field is important chiefly as a convergence factor; its magnitude is small, and does not enter explicitly in most cases. We shall therefore, for simplicity, assume that it may also be multiplied by ϕ, and the corrected formula for the energy levels becomes

$$E^* = [\Sigma_\lambda(N_\lambda^+ + N_\lambda^- + 1)\mu_\lambda][1-(2NS)^{-1}\Sigma_\lambda\{(N_\lambda^+ + N_\lambda^- + 1)A_\lambda/\mu_\lambda - 1\}] + \Sigma_\lambda(N_\lambda^+ - N_\lambda^-)g\beta H_z, \quad \ldots\ldots(27)$$

where the sum is again over the half space of $\mathbf{K}_\lambda$.

Use of this result in place of our previous equation (1) should lead to better estimates of the properties of the system, and there seems no reason why it should not apply over a wide range of temperature, up to near the Curie point. The total number of levels is properly restricted by the limit on ϕ. We may note that the domain stability condition, $\mu_\lambda > 0$, is also required in order that the sum in (26) should be finite, although when the sum is replaced by an integral this may not be significant numerically.

§ 5. THE GROUND STATE

As before, this is the state in which each $N_\lambda^\pm$ is zero. We shall be left only with the zero-point motion, but this will be effective in making ϕ differ from unity. Indeed, for the linear chain, the integral corresponding to (26) does not converge, and the correction is indeterminate; the agreement of the uncorrected result with that of Hulthén (1938), previously noted, must be fortuitous. But in the other cases we need only multiply γ by the zero-point value ϕ_0 of ϕ to correct the table given in I. The correction depends on the actual value of S, and may be written

$$\gamma' = \phi_0\gamma = (1 - n/2S)\gamma \quad \ldots\ldots(28)$$

where $n = 0{\cdot}195$, $0{\cdot}075$, $0{\cdot}078$ for the plane quadratic, simple cubic and body-centred cubic lattices respectively.

We may also obtain an expression for the average value of $|S_l^{(z)}|$. The results are identical with the formulae deduced by Anderson (1951).

§ 6. CORRECTION TO $\chi_\perp$

Reference to (9) and (10), and comparison with I (8), etc., will show that we have dropped a factor $\sqrt{\phi}$ in $\mathscr{H}'$. There should also be a factor ϕ in the perturbed Hamiltonian $\mathscr{H}$, and since $\mathscr{H}'$ is squared in calculating the second-order perturbation, these factors exactly cancel out. This is the case whatever the state of occupation of the levels, i.e. whatever the temperature. Thus (16) should be true at all temperatures—or at least until the parallel order has entirely broken down. Even taking into account the correlation effects, this is still true.

§ 7. CORRECTION TO $\chi_\parallel$

Although (27) is probably a good description of the distribution of the energy levels, it is impossible to put this into an expression for the partition function and perform the summations exactly. We must approximate, as follows. In ϕ, instead of N_λ we write the thermodynamic average value of N_λ at the given temperature and in zero field. Thus, in the partition function,

ϕ is a constant for the summations over N_λ, and the average value of N_λ at a given temperature is obtained from

$$(N_\lambda+\tfrac{1}{2})_{\mathrm{av}} = -\frac{\boldsymbol{k}T}{\phi}\frac{\partial}{\partial\mu_\lambda}\Sigma_{N_\lambda}\exp-\{\Sigma_\lambda(N_\lambda+\tfrac{1}{2})\phi\mu_\lambda/\boldsymbol{k}T\} = \tfrac{1}{2}\coth(\phi\mu_\lambda/2\boldsymbol{k}T). \quad .(29)$$

This is the value of $N_\lambda+\frac{1}{2}$ to be put into (26), so that we obtain the implicit equation

$$\phi = 1-(2SN)^{-1}\Sigma_\lambda\tfrac{1}{2}\{\coth(\phi\mu_\lambda/2\boldsymbol{k}T)A_\lambda/\mu_\lambda-1\} \quad \ldots\ldots(30)$$

from which ϕ is obtained as a function of T. (This device for approximating to the partition function is not altogether satisfactory when it comes to calculating thermal properties, because the free energy and internal energy obtained by the usual thermodynamic formulae do not quite obey the Gibbs–Helmholtz relation, but the difficulty only arises when we wish to differentiate with respect to T.)

The determination of the parallel susceptibility can now be done without difficulty, and we obtain, in place of (6),

$$\chi_{\parallel}^{*} = \frac{N}{V}\frac{1}{64\pi^3}\frac{g^2\beta^2}{\boldsymbol{k}T}\int\int_{-\pi}^{\pi}\int\left\{\operatorname{cosech}^2\left(\frac{\phi\mu_\lambda+g\beta H_z}{2\boldsymbol{k}T}\right)\right.$$
$$\left.+\operatorname{cosech}^2\left(\frac{\phi\mu_\lambda-g\beta H_z}{2\boldsymbol{k}T}\right)\right\}d\mathbf{K}_\lambda. \quad \ldots\ldots(31)$$

It is usual to assume that the total susceptibility χ of a substance containing randomly oriented domains is given by $\chi=\frac{1}{3}\chi_{\parallel}^{*}+\frac{2}{3}\chi_{\perp}$ (Van Vleck 1941), and in fig. 2 we have plotted $\chi/S\chi_0$ as a function of $\boldsymbol{k}T/2S|J|$ for the simple cubic

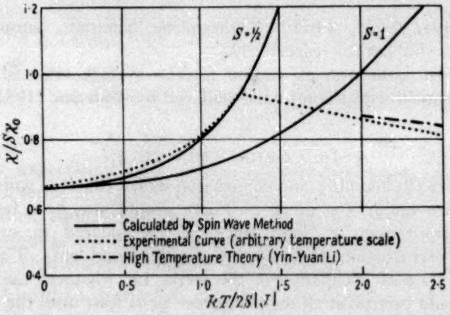


Fig. 2. Temperature variation of susceptibility for simple cubic lattice.

lattice with various values of S. For comparison a typical experimental curve (Van Vleck 1941) is shown. The comparison cannot be made precisely since we have no means of deciding the exact value of the critical temperature by our method. The following points may be noted:

(*a*) The shape of the theoretical curves agrees with experiment over a wide range of temperature.

(*b*) As $T\to 0$, $\chi_{\parallel}^{*}\to\dfrac{1}{6\sqrt{3}}\dfrac{1}{\phi_0^2}\left(\dfrac{\boldsymbol{k}T}{2S|J|}\right)^2\chi_0$.

(*c*) The susceptibility rises rapidly as the temperature approaches a definite limit, which may be taken as the upper limit for an estimate of the Curie temperature. Thus, when the spin is $\frac{1}{2}$, this limiting temperature is given by $\boldsymbol{k}T/2S|J|=1{\cdot}7$. This lies rather below the estimates of other theories, the lowest of which is that of Yin-Yuan Li (1951), who found $\boldsymbol{k}T_c/2S|J|=2{\cdot}0$. He also found that the corresponding susceptibility χ_c is given by $\chi_c=0{\cdot}871\,S\chi_0$, and we have plotted in fig. 2 the variation he found for the susceptibility above that point. It is possible that his method, which is an accurate quantum-mechanical treatment of the cluster of atoms surrounding a given site, gives very good values for the susceptibility, which depends chiefly on the short-range order above the Curie point, but that it is not accurate in fixing the critical temperature itself, since this depends on cooperative phenomena, involving the whole lattice.

(*d*) It is suggested that if his calculations could be extended below this point, ignoring the possibility of the existence of an ordered state, they would define a curve which would intersect our curve at a point very close to the true critical temperature. Indeed, a rough graphical extrapolation of his published curve suggests that it should cut at a value of $\chi/S\chi_0$ somewhat below unity; this would help to explain the observed ratio of $\chi(T=0)$ to χ_c, which is always larger than the value 2/3 predicted by internal field theories. The reduction in the value of T_c (which appears to lie near $1{\cdot}2\times 2S|J|/\boldsymbol{k}$) may also explain the large experimental ratios of θ/T_c, where θ is the constant in the approximate high-temperature Curie–Weiss law, $\chi=\chi'/(T+\theta)$ (see, for example, Anderson 1950). The precise values of these ratios probably depend, however, on the spins, range of interaction, and lattice arrangement of the atoms.

§8. CONCLUSION

Throughout this work we have not attempted to discuss any particular type of system in detail, except the simple cubic lattice, which has served as a convenient model and example. We have tried, rather, to construct a theoretical framework of sufficient generality to cover a wide variety of possibilities. Application of the method to more complex lattices is at present under investigation, but the following summarizes the range of application of the theory: (*a*) Any type of lattice with one magnetic ion per unit cell may be studied. (*b*) Any type of interaction Hamiltonian may be used, and the range of the interaction forces need not be restricted to a single lattice spacing (although for long-range forces, involving many atoms, the computation may become too laborious). (*c*) The method is not restricted to vanishingly small external fields, as long as due account is taken of anisotropy energy in maintaining stable domains. (*d*) The calculations are reliable up to temperatures approaching the critical temperature, and because we have used a representation in terms of operators with exact commutation relations it is possible to make further corrections to the energy levels by simple perturbation methods.

ACKNOWLEDGMENTS

Part of this work was done during the tenure of a maintenance grant from the Department of Scientific and Industrial Research. It is a pleasure to thank Professor G. S. Rushbrooke for his encouragement and advice, and Dr. K. W. H. Stevens for numerous valuable discussions. The suggestions of the referees have also proved very helpful.

APPENDIX

We wish to calculate the expectation value of $(a_l{}^*a_la_m{}^*a_m)_{\text{av}}$, where l and m are neighbours and the average is over all sites in the lattice. Thus

$$(a_l{}^*a_la_m{}^*a_m)_{\text{av}} = N^{-1}\,\Sigma_l a_l{}^*a_la^*_{l+1}a_{l+1},$$

where the $(l+1)$th site is the neighbour of the lth, and distant vector $\mathbf{a}$ from it. In terms of the Fourier transform operators

$$\begin{aligned}(a_l{}^*a_la_m{}^*a_m)_{\text{av}} &\\ &= N^{-3}\,\Sigma_{l\lambda\mu\nu\sigma}\alpha_\lambda{}^*\alpha_\mu\alpha_\nu{}^*\alpha_\sigma \exp i\{\mathbf{R}_l.\mathbf{K}_\lambda - \mathbf{R}_l.\mathbf{K}_\mu + (\mathbf{R}_l+\mathbf{a}).\mathbf{K}_\nu - (\mathbf{R}_l+\mathbf{a}).\mathbf{K}_\sigma\}\\ &= N^{-2}\,\Sigma_{\lambda\mu\nu\sigma}\alpha_\lambda{}^*\alpha_\mu\alpha_\nu{}^*\alpha_\sigma \exp\{i\mathbf{a}.(\mathbf{K}_\nu - \mathbf{K}_\sigma)\}\delta(\lambda-\mu+\nu-\sigma).\end{aligned}$$

This operator is to be evaluated in a given state $|N_\lambda\rangle$. From the properties of the operators the expectation value vanishes unless $\lambda=\mu$, $\nu=\sigma$, or $\lambda=-\nu$, $\mu=-\sigma$, or $\lambda=\sigma$, $\mu=\nu$. But consider, for example, this last case:

$$\begin{aligned}\langle N_\lambda, N_\mu |\, N^{-2}\,\Sigma_{\lambda\mu}\alpha_\lambda{}^*\alpha_\lambda\alpha_\mu{}^*\alpha_\mu \exp\{i\mathbf{a}.(\mathbf{K}_\lambda - \mathbf{K}_\mu)\}\,| N_\lambda, N_\mu\rangle &\\ = \left|\frac{1}{N}\Sigma_\lambda\left\{(N_\lambda+\tfrac{1}{2})\frac{A_\lambda}{\mu_\lambda} - \frac{1}{2}\right\}\exp(i\mathbf{a}.\mathbf{K}_\lambda)\right|^2.\end{aligned}$$

In the state in which $N_\lambda+\frac{1}{2}$ is given by the average (28) this is zero, since there are always pairs of terms for which A_λ and μ_λ are of the same but $\exp(i\mathbf{a}.\mathbf{K}_\lambda)$ of opposite sign. Since the correlation factor is only a small correction to the Hamiltonian in any case, the most probable distribution of occupation numbers will be that of (28), and there will be no contribution from these terms. The only term which does not thus vanish is that in which $\lambda=\mu$, $\nu=\sigma$, and we have, immediately,

$$\langle a_l{}^*a_la_m{}^*a_m\rangle_{\text{av}} = N^{-2}\,\Sigma_{\lambda\mu}\langle\alpha_\lambda{}^*\alpha_\lambda\rangle\,\langle\alpha_\mu{}^*\alpha_\mu\rangle = (\langle a_l{}^*a_l\rangle)^2.$$

REFERENCES

ANDERSON, P. W., 1950, *Phys. Rev.*, **79**, 705; 1951, *An Approximate Quantum Theory of the Antiferromagnetic Ground State* (Bell Telephone Laboratories Report).

HOLSTEIN, T., and PRIMAKOFF, H., 1940, *Phys. Rev.*, **58**, 1098.

HULTHÉN, L., 1936, *Proc. Amst. Acad. Sci.*, **39**, 190; 1938, *Ark. Mat. Astr. Fys.*, **26A**, No. 11.

KITTEL, C., 1951, *Phys. Rev.*, **82**, 565 (L).

NAGAMIYA, T., 1951, *Prog. Theor. Phys.*, **6**, 342.

VAN VLECK, J. H., 1941, *J. Chem. Phys.*, **9**, 85.

YIN-YUAN LI, 1951, *Phys. Rev.*, **84**, 721.

ZIMAN, J. M., 1952, *Proc. Phys. Soc.* A, **65**, 540.

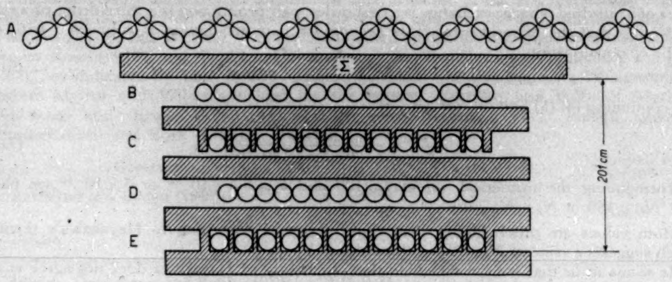
LETTERS TO THE EDITOR

An Anticoincidence Experiment on Cosmic Rays at a Depth of 50 Metres Water Equivalent

The existence of a neutral component of cosmic rays underground has been suggested several times in the past but always rejected. Quite recently it has been reviewed again, in a new form, by Hayakawa (1952) with the aim of explaining the pairs of penetrating particles, which, according to various authors (Braddick *et al.* 1951, George and Trent 1949) are produced underground by the incident charged particles with a cross section σ_{exp} of about 5×10^{-29} cm^2/nucleon.

We have recently investigated this problem and obtained results which seem to indicate that the cross section of the phenomena described above is appreciably smaller ($\sigma_{\text{exp}} \sim 5 \times 10^{-30}$ cm²/nucleon) than was thought before (Amaldi *et al.* 1952). If our conclusion is definitely proved it will no longer be necessary to invoke the existence of particles other than μ mesons in order to explain the observed pair production.

However, at the end of his paper Hayakawa suggests, as a test of his theory, an anti-coincidence experiment which happens to have been made by us during our work underground. We report here only on this point while a more extensive discussion of our results will appear in *Il Nuovo Cimento*. The figure shows our experimental set-up. We consider in this letter only the fourfold coincidences C_4 between the four sets of counters BCDE (all counters of each set are in parallel; between the various counter sets there is a total of 15 cm Pb) and the fivefold coincidences C_5, counted simultaneously, between the four preceding sets and the counters A all in parallel. Measurements have been made with and without the layer of Pb Σ whose thickness Δ_Σ was either 5 or 7·5 cm.



Experimental set up of counters (not to scale).

Assuming with Hayakawa that underground there are, besides charged particles, also neutral particles which can undergo a charge exchange process with cross section σ_0, we have

without the lead $$C_4(0) = N_c G_1, \quad C_5(0) = \eta_A N_c G_1, \qquad \ldots\ldots (1)$$

with the lead Σ $$C_4(\Delta_\Sigma) = [N_c \epsilon + N_{K^0} \sigma_0 \boldsymbol{N} \Delta_\Sigma]\, G_1, \quad C_5(\Delta_\Sigma) = \epsilon \eta_A N_c G_1 \qquad \ldots\ldots (2)$$

where G_1 is a geometrical factor of the telescope, N_c and N_{K^0} the numbers of charged and neutral particles incident in the vertical direction per cm² and steradian, η_A the efficiency of the set of counter A, ϵ the absorption of Σ and $\boldsymbol{N}$ Avogadro's number.

Combining (1) with (2) we get

$$\frac{N_{K^0}\sigma_0}{N_c} = \frac{1}{\Delta_\Sigma \boldsymbol{N}} \frac{1}{C_4(0)} \left\{ [C_4(\Delta_\Sigma) - C_5(\Delta_\Sigma)] - \frac{C_5(\Delta_\Sigma)}{C_5(0)} [C_4(0) - C_5(0)] \right\}. \qquad \ldots\ldots (3)$$

Equation (3) is written in terms of the differences between the fourfold and the fivefold coincidence rates in order to take advantage of the fact that with our coincidence circuits the fivefold coincidences were not statistically independent of the fourfold coincidences.

Δ_Σ(g/cm²)	t (min)	C_4	c_4 (counts/min)	C_5	c_4 (counts/min)	$c_4 - c_5$
0	21·803	279·188	12·805±0·024	269·798	12·374±0·024	0·431±0·0045
57	21·018	265·458	12·630±0·024	256·728	12·215±0·024	0·415±0·0044
85	18·203	226·433	12·439±0·026	218·786	12·019±0·026	0·420±0·0049

Our results are given in the table. Introducing the appropriate values in eqn. (3) we get

for $\Delta_\Sigma = 57$ g/cm², $N_{K^0}\sigma_0/N_c = (-2{\cdot}5 \pm 1{\cdot}5) \times 10^{-29}$ cm²/nucleon,

for $\Delta_\Sigma = 85$ g/cm², $N_{K^0}\sigma_0/N_c = (1{\cdot}5 \pm 1) \times 10^{-29}$ cm²/nucleon.

These two values are equal inside the experimental errors and therefore we can take their mean value which is

$$\frac{N_{\kappa^0}\sigma_0}{N_c}=(-1\pm1{\cdot}5)\times10^{-29}\ \mathrm{cm}^2/\mathrm{nucleon}. \qquad \ldots\ldots (4)$$

Now, according to Hayakawa (1952)

$$N_c=N_\mu+N_{\kappa\pm} \quad \text{and} \quad \sigma_0=f\sigma_\Lambda, \qquad \ldots\ldots (5)$$

where N_μ and $N_{\kappa\pm}$ are the numbers of μ mesons and of $\kappa\pm$ mesons, σ_Λ is the cross section for production of pairs of penetrating particles by $\kappa\pm$ mesons, and f a numerical factor of the order of a hundred (charge exchange is a first order, pair production is a third order process).

Noting that the experimental cross section for pair production is connected with σ_Λ by the obvious relation

$$\sigma_{\mathrm{exp}}=\frac{N_{\kappa\pm}}{N_\mu+N_{\kappa\pm}}\sigma_\Lambda \qquad \ldots\ldots (6)$$

and combining (4) (5) and (6), we get

$$\frac{N_{\kappa\pm}}{N_{\kappa^0}}=\frac{f\,\sigma_{\mathrm{exp}}}{(-1\pm1{\cdot}5)\times10^{-29}}. \qquad \ldots\ldots. (7)$$

Introducing the numerical values $f\simeq100$ and $\sigma_{\mathrm{exp}}=5\times10^{-29}$ or 5×10^{-30} we have $N_{\kappa\pm}/N_{\kappa^0}\gtrsim150$ or $N_{\kappa\pm}/N_{\kappa^0}\gtrsim15$.

Both values are much higher than those expected according to Hayakawa's theory, which suggests a value of $N_{\kappa\pm}/N_{\kappa^0}$ of the order of 1.

It seems to us that we can conclude that the theory of Hayakawa does not agree at all with our anticoincidence experiment, whatever is the value of the cross section for pair production by cosmic rays underground.

Istituto di Fisica dell'Università, — E. Amaldi.
Centro di Studio per la Fisica Nucleare del C.N.R., — C. Castagnoli.
Roma. — S. Sciuti.

Istituto di Fisica dell 'Università — A. Gigli.
Pavia.
15th April 1952.

Amaldi, E., Castagnoli, C., Gigli, A., and Sciuti, S., 1952, *Nuovo Cim.* (in the press).
Braddick, H. J., Nash, W. F., and Wolfendale, A. W., 1951, *Phil. Mag.*, **42,** 1277.
George, E. P., and Trent, P., 1949, *Nature, Lond.*, **164,** 839.
Hayakawa, S., 1952, *Proc. Phys. Soc.* A, **65,** 215.

Note on the Differential Momentum Spectrum of Cosmic-Ray Protons at Sea Level

Rossi (1948) has used the data of Rochester and Bound (1940) to determine one point of the differential momentum spectrum of cosmic-ray protons at sea level. Due to a misprint in the text of Rossi's article, several authors (e.g. Mylroi and Wilson 1951, Rochester and Rosser 1951, and Puppi and Dallaporta 1951) have given an incorrect value for this spectral point. The misprint in the text of Rossi's article states that the number of protons within a range interval of 2 cm of lead is 5×10^{-7} measured in units of cm^{-2} sec^{-1} sterad^{-1}. As seen in the table accompanying the text, this figure actually is the value of the differential range spectrum of protons in g^{-1} sec^{-1} sterad^{-1}. Thus the flux of protons through Rochester and Bound's apparatus is $8{\cdot}6\times10^{-6}$ cm^{-2} sec^{-1} sterad^{-1}.

Taking into account the wall thicknesses of the counters, cloud chamber and housing around the apparatus, all protons with momentum in the range $0{\cdot}34-0{\cdot}53\times10^9$ ev/c would be stopped in the 2 cm lead block. The flux of protons in this momentum range corresponds to a vertical intensity of $I_v=4{\cdot}5\pm1{\cdot}6\times10^{-8}$ cm^{-2} sec^{-1} sterad^{-1} (Mev/c)$^{-1}$.

Because the purpose of the experiment of Rochester and Bound was to measure the attenuation of protons in lead rather than to determine the intensity of protons, a very stringent criterion was placed upon those tracks which were accepted as protons. The eight particles considered were probably all greater than about four times minimum ionization. The momentum of a proton with this ionization is only $0{\cdot}40 \times 10^9$ ev/c, and not the upper limit of $0{\cdot}53 \times 10^9$ ev/c used in the above calculation, which corresponds to 2·5 times minimum ionization. If now we take the momentum interval for counted protons to be 0·34 to $0{\cdot}40 \times 10^9$ ev/c instead of 0·34 to $0{\cdot}53 \times 10^9$ ev/c, we obtain a corrected value of the spectrum $I_V = 14 \pm 5 \times 10^{-8}$ cm^{-2} sec^{-1} sterad^{-1} (Mev/c)$^{-1}$.

The table gives a summary of the present data available in the low momentum region of the differential spectrum for protons. Two sets of values of the spectrum are given for the data of Mylroi and Wilson. The first is obtained from the differential meson spectrum given by Rossi (1948), and the second by assuming that Rossi's meson spectrum is too low by 20%, as indicated by York (1952). It is to be noted that this improves the agreement between Mylroi and Wilson's values for the proton spectrum and those of Merkle, Goldwasser and Brode (1950), Goldwasser and Merkle (1951), Ballam (private communication) and York (1952).

Observer	Momentum interval ($\times 10^{-9}$ ev/c)	I_V ($\times 10^8$)	Corrected I_V ($\times 10^8$)
Rochester and Bound (1940)	0·34–0·53	4·5±1·6	—
	0·34–0·40	—	14±5
Ballam	0·42–0·91	30±8	—
	0·51–0·96	31±14	—
Merkle, Goldwasser and Brode (1950)	0·48–0·88	23±3	—
Goldwasser and Merkle (1951)	0·78–1·10	20±5	—
Mylroi and Wilson (1951)	0·53–0·71	16·8±4·4	20·1±5·3
	0·71–0·93	8·9±1·6	10·7±1·9
	0·93–1·35	5·5±0·9	6·6±1·1
York (1952)	0·55–0·75	28±3	—
	0·75–0·88	10·7±0·8	—

The author is indebted to Professor Rossi for pointing out the inconsistency between the text and table in his article and to Dr. G. D. Rochester for supplying the original data from the experiment of Rochester and Bound. The author would like to express his thanks to Dr. Rochester and Professor R. B. Brode for several stimulating conversations and for checking the calculations used in the table. He would also like to thank Professor P. M. S. Blackett for extending the facilities of his laboratory during the author's stay in Manchester and the United States Educational Commission in the United Kingdom for financial support.

The Physical Laboratories,
University of Manchester.
9th May 1952.

CARL M. YORK.

GOLDWASSER, E. L., and MERKLE, T. C., 1951, *Phys. Rev.*, **83**, 43.
MERKLE, T. C., GOLDWASSER, E. L., and BRODE, R. B., 1950, *Phys. Rev.*, **79**, 926.
MYLROI, M. G., and WILSON, J. G., 1951, *Proc. Phys. Soc.* A, **64**, 404.
PUPPI, G., and DALLAPORTA, N., 1951, *Progress in Cosmic Ray Physics* (Amsterdam: North Holland), p. 317.
ROCHESTER, G. D., and BOUND, M., 1940, *Nature, Lond.*, **146**, 745.
ROCHESTER, G. D., and ROSSER, W. G. V., 1951, *Rep. Prog. Phys.*, **14**, 227 (London: Physical Society).
ROSSI, B., 1948, *Rev. Mod. Phys.*, **20**, 537.
YORK, C. M., 1952, *Phys. Rev.*, **85**, 998.

Paramagnetic Resonance in Ferric Rubidium Sulphate Alum

Ferric ammonium sulphate alum, in which the ferric ion is in a ^{6}S state, has been extensively used in adiabatic demagnetization experiments. The sextet level is split by the crystalline field; if the field has cubic symmetry a doublet and a quadruplet remain, whose separation, according to specific heat measurements (Cooke 1949) is 0·14 cm^{-1}. Bagguley *et al.* (1948) found, however, that paramagnetic resonance experiments indicated a splitting, assuming a cubic field, of less than 0·1 cm^{-1}; they suggested that a trigonal field was present which further splits the quadruplet into two doublets. In two salts of manganese, where the Mn^{2+} ion is isoelectronic with Fe^{3+}, the trigonal splitting is much larger than the cubic splitting (Bleaney 1950, Bleaney and Ingram 1951).

Various attempts (Bijl 1950, Wiedner *et al.* 1949, Ubbink *et al.* 1951) to clarify the position in the iron alums by the use of diluted salts have not been completely successful because the side lines are unduly broad, presumably because the splitting of the levels by the crystalline field is not uniform throughout the crystal. Meyer (1951) tried to explain the observed spectra using a combined cubic and trigonal field similar to that found to be successful in the manganese salts, but some discrepancies remained, though no exact test of the theory was possible owing to lack of experimental resolution.

We have found that high resolution can be obtained with diluted ferric rubidium sulphate alum. The results show that the cubic axes of the ferric ion are not identical with those of the crystal, but are rotated by an angle of $7\frac{1}{2}\pm\frac{1}{2}°$ about the trigonal axis. The splitting can be interpreted using the Hamiltonian for manganese (Bleaney and Ingram 1951) with an additional trigonal term corresponding to the spherical harmonic Y_4^0, whose magnitude is denoted by the parameter F. In a strong field making an angle θ with the trigonal axis, the spectrum consists of five lines, whose positions are given by the formulae (correct only to the first order)

$$h\nu=g\beta H\pm\{2pa+2D(3\cos^2\theta-1)+\tfrac{1}{6}F(35\cos^4\theta-30\cos^2\theta+3)\}$$
$$h\nu=g\beta H\pm\{-\tfrac{5}{2}pa+D(3\cos^2\theta-1)-\tfrac{5}{24}F(35\cos^4\theta-30\cos^2\theta+3)\}$$
$$h\nu=g\beta H$$

The values of the constants are as follows (a, D, F in cm^{-1}) :

Temp. (°K)	g	a	D	F
90	2·004 ± 0·003	−0·0134	+0·0022	−0·0003
20	2·004 ± 0·001	−0·0134	+0·0031	−0·0003

The relative signs of the constants are found from fitting the spectrum, the actual signs from the varying relative intensity of the lines owing to their unequal populations at low temperatures.

The rotation of the cubic axes is deduced from a detailed examination of the spectrum at intervals of 5° in the (100) and (110) planes, and is slightly smaller than that ($9\frac{1}{2}°$) found by Lipson (1935) from x-ray analysis of aluminium rubidium alum. The excellent agreement with the measurements that can be obtained with the values of a, D and F given above is illustrated by the following table. This gives the splitting between the central line and the side lines at 20 °K, second-order displacements being eliminated by taking the mean of corresponding lines on either side.

Direction	Multiplicity	Experimental splitting (gauss)		Calculated splitting (gauss)
(100)	Quadruplet	264		262
	,,	329		327
(111)	Singlet	167		169
	,,	320		319
	Triplet	233		235
	,,	124		125
(110)	Doublet	123	unresolved	128
	,,	123		125
	,,	45		43
	,,	unresolved from central line		4

In zero field the levels form three doublets, calculated to lie at 0, 0·0026° and 0·0633°K respectively. The corresponding contribution to the magnetic specific heat tail is $CT^2/R=2a^2+56D^2/9+14F^2/27-28aF/27=8{\cdot}58\times10^{-4}$ (°K)^2.

Preliminary measurements on ferric potassium selenate alum and ferric methylamine sulphate alum indicate that a is about the same as in the rubidium alum, but the values of D are roughly 0·01 cm^{-1} and 0·19 cm^{-1} respectively, the sign in each case being the same as that of a.

Clarendon Laboratory,
Oxford.
13th May 1952.

B. Bleaney.
R. S. Trenam.

Bagguley, D. M. S., Bleaney, B., Griffiths, J. H. E., Penrose, R. P., and Plumpton, B. I., 1948, *Proc. Phys. Soc.*, **61**, 542.
Bijl, D., 1950, *Thesis*, Leiden.
Bleaney, B., 1950, *Phys. Rev.*, **78**, 214.
Bleaney, B., and Ingram, D. J. E., 1951, *Proc. Roy. Soc.* A, **205**, 336.
Cooke, A. H., 1949, *Proc. Phys. Soc.* A, **62**, 269.
Lipson, H., 1935, *Proc. Roy. Soc.* A, **151**, 347.
Meyer, P. H. E., 1951, *Physica*, **17**, 899.
Ubbink, J., Poulis, J. A., and Gorter, C. J., 1951, *Physica*, **17**, 213.
Weidner, R. T., Weiss, P. R., Whitmer, C. A., and Blosser, D. R., 1949, *Phys. Rev.*, **76**, 1727.

A Note on the Theory of Conduction in Metals

The object of the present note is to draw attention to a feature of the theory of metallic conduction which appears to have been overlooked. In the usual version of the theory (see, for example, Wilson 1936) it is assumed that electrons can be scattered by lattice vibrations with all wave numbers $|\mathbf{q}|\leqslant q_0$, where q_0 corresponds to the cut-off frequency of the Debye spectrum. However, in any collision involving a (quasi-free) electron with wave vector $\mathbf{k}$ and a lattice wave $\mathbf{q}$, the laws of conservation of energy and momentum require that $|\mathbf{k}|=|\mathbf{k}\pm\mathbf{q}|$, and thus an electron in the state $\mathbf{k}$ can only interact with lattice waves for which $|\mathbf{q}|\leqslant 2|\mathbf{k}|$. This fact is well known in the theory of semiconductors, and has also been taken into account in Fröhlich's theory of superconductivity (Fröhlich 1950); but its consequences for the theory of normal metals do not seem to have been noticed. In a metal it is only necessary to consider electrons with wave vectors $|\mathbf{k}|=K_0$ corresponding to the surface of the Fermi distribution. It follows that, if $2K_0<q_0$, the upper limit of integration in the collision operator is $2K_0$ and not q_0. A number of interesting consequences follow.

(*a*) If $2K_0<q_0$ the usual expression for the time of relaxation at high temperatures (Wilson 1936, p. 208) must be increased by a factor $(q_0/2K_0)^4$. Since $K_0/q_0=(\frac{1}{2}N_a)^{1/3}$, where N_a is the number of conduction electrons per atom, the condition $2K_0<q_0$ is equivalent to $N_a<\frac{1}{4}$, and the factor $(q_0/2K_0)^4$ equals $(4N_a)^{-4/3}$. The high-temperature electrical and thermal conductivities are to be multiplied by the same factor.

(*b*) The usual interpolation formulae for the temperature variation of the electrical and electronic thermal conductivities of metals (discussed, for example, by Makinson 1938) involve the parameter $D/\zeta=q_0^2/2K_0^2=2^{-1/3}N_a^{-2/3}$. These formulae are clearly only valid if $D/\zeta\leqslant 2$, and so do not apply to metals in which the number of conduction electrons is small; in particular, Makinson's discussion of the thermal conductivity of bismuth is not valid.

(*c*) The modified interpolation formulae which hold when $D/\zeta>2$ are easily obtained by the usual methods (Wilson 1937, Sondheimer 1950). In the notation used by these authors it is found that, for an ideally pure metal,

$$\frac{1}{\sigma_1}=\frac{3\pi\boldsymbol{h}D}{4\boldsymbol{e}^2\zeta^3\Lambda}\left(\frac{T}{\Theta}\right)^5\mathcal{J}_5\left(\frac{\Theta'}{T}\right), \qquad \ldots\ldots(1)$$

$$\frac{1}{\kappa_1}=\frac{27\boldsymbol{h}}{8\pi^3\boldsymbol{k}^2\zeta^2\Lambda\Theta}\left(\frac{T}{\Theta}\right)^2\left[\mathcal{J}_5\left(\frac{\Theta'}{T}\right)+\frac{D}{\zeta}\left(\frac{\Theta}{T}\right)^2\left\{\frac{2\pi^2}{3}\mathcal{J}_5\left(\frac{\Theta'}{T}\right)-\frac{1}{3}\mathcal{J}_7\left(\frac{\Theta'}{T}\right)\right\}\right], \qquad \ldots\ldots(2)$$

where $\Theta' = \sqrt{(2\zeta/D)}\Theta$. It follows that the temperature variation of σ and κ is *qualitatively* unchanged; the values of the conductivities at sufficiently low temperatures are also not affected, as is obvious since only long lattice waves are then excited in any case. The ratio of the high- to the low-temperature conductivities is thus increased by the factor $(D/2\zeta)^2 = (4N_a)^{-4/3}$. Further, the transition from 'high' to 'low' temperature behaviour takes place at a temperature Θ' which is less than the Debye temperature Θ.

The present note has been written mainly in order to warn against the use of the usual theory in cases where it does not apply. The new formulae do not, unfortunately, have more than qualitative significance, since the metals in which the number of conduction electrons is small (the most popular example of which is bismuth) are just those in which the simplifying assumptions of the theory will be furthest from the truth.

It is perhaps, however, worth while considering briefly the implications of the new theory for the properties of bismuth. In this metal N_a is of the order of 10^{-3}, so that $D/\zeta \sim 80$ and $q_0/2K_0 \sim 6$. (Makinson (1938) adopted a much higher value of N_a, apparently confusing the actual number of free electrons in a metal with the 'effective' number in the sense defined by Mott and Jones (1936).) The most striking result of the new theory is that the time of relaxation, and with it the free path, of the electrons in bismuth are likely to be much larger than the corresponding quantities in 'good' conductors. A quantitative estimate of the absolute values would, of course, require a much more refined theory, in view of the many uncertain parameters involved and the possible effect of additional scattering processes (inter-band transitions, 'Umklapprozesse') which have been neglected. It is remarkable, however, that experiments on thin films and wires of bismuth (Eucken and Förster 1934, Justi, Kohler and Lautz 1951) indicate that the free path at room temperatures is of the order of 10^{-4} cm, which is about 20 times larger than the free path in silver. If the present suggestion is correct, this is simply due to the fact that slow electrons are prevented by the conservation laws from interacting with any but the longest lattice vibrations. The fact that bismuth has, in spite of this, a small electrical conductivity is, of course, due to the fact that the small number of conduction electrons outweighs the effect of the long free path.

The present theory further predicts that the electrical resistance of bismuth should vary linearly with temperature down to temperatures of the order of 10° K, which does not agree with observation. This indicates the grossly oversimplified nature of the present model. The thermal conductivity should be independent of temperature, and the Lorenz number should have its normal value, down to temperatures of the same order. It is, unfortunately, difficult to test these predictions owing to the presence in bismuth of a large lattice component of thermal conductivity.

The considerations outlined above also have a bearing on the interpretation of recent experiments on the thermal conductivity of monovalent metals. This question is taken up in the following note.

Department of Mathematics,
Imperial College, London.
2nd May 1952.

E. H. SONDHEIMER.

EUCKEN, A., and FÖRSTER, F., 1934, *Nachr. Ges. Wiss. Göttingen*, **1**, 43.
FRÖHLICH, H., 1950, *Phys. Rev.*, **79**, 845.
JUSTI, E., KOHLER, M., and LAUTZ, G., 1951, *Z. Naturforsch.*, **6a**, 456.
MAKINSON, R. E. B., 1938, *Proc. Camb. Phil. Soc.*, **34**, 474.
MOTT, N. F., and JONES, H., 1936, *The Theory of the Properties of Metals and Alloys* (Oxford: Clarendon Press), p. 97.
SONDHEIMER, E. H., 1950, *Proc. Roy. Soc.* A, **203**, 75.
WILSON, A. H., 1936, *The Theory of Metals* (Cambridge: University Press), chapter VI; 1937, *Proc. Camb. Phil. Soc.*, **33**, 371.

The Thermal Conductivity of Metals at Low Temperatures

Recent experiments on the thermal conductivity of various metals at low temperatures (Hulm 1950, Berman and MacDonald 1951, 1952) have made it possible to check the predictions of the theory (Makinson 1938, Sondheimer 1950) in much greater detail than hitherto. In the present note we wish to make some comments on the comparison between theory and experiment.

The electronic thermal conductivity κ at sufficiently low temperatures is given by the formula

$$1/\kappa=\alpha T^2+\beta/T, \qquad \ldots\ldots(1)$$

where the first term represents the effect of the lattice vibrations and the second term that of impurities and other static lattice defects. In considering the theoretical value of α (following Hulm), it must, however, be remembered that the theoretical formulae are different according as $D/\zeta \gtrless 2$. (For the significance of this parameter see Makinson 1938, and the preceding note, Sondheimer 1952.) In fact we have

$$\alpha=6\zeta\mathcal{J}_5(\infty)/\pi^2 D\kappa_\infty\Theta^2 \qquad \text{if } D/\zeta\leqslant 2, \qquad \ldots\ldots(2\,a)$$

$$\alpha=3D\mathcal{J}_5(\infty)/2\pi^2\zeta\kappa_\infty\Theta^2 \qquad \text{if } D/\zeta>2, \qquad \ldots\ldots(2\,b)$$

where κ_∞ is the thermal conductivity at high temperatures and the notation is otherwise the same as Makinson's. If α is regarded as a function of D/ζ it has a minimum value $\alpha_{\text{min}}=3\mathcal{J}_5(\infty)/\pi^2\kappa_\infty\Theta^2=37{\cdot}8/\kappa_\infty\Theta^2$, which occurs when $D/\zeta=2$. The observed values of α are in all cases rather smaller than this minimum value; some typical examples are shown in the following table. (The values of Θ have been taken equal to those which give the best fit for the electrical resistance.)

	Na($\Theta=202$)	Cu($\Theta=330$)	Pb($\Theta=88$)
$10^4\alpha_{\text{min}}$	7·1	0·90	140
$10^4\alpha$ (observed)	3·8	0·23	22

(κ is measured in watts/cm/°K)

Thus no value at all of D/ζ will fit the observations. Hulm, who assumed (with Makinson) that eqn. (2 *a*) was valid for all values of D/ζ, concluded that the experiments appear to indicate an abnormally large value of D/ζ (and hence a small number of conduction electrons) in the metals studied; we now see, however, that, whatever the explanation of the discrepancy, this interpretation is not admissible.

The theoretical value of D/ζ in monovalent metals is $2^{-1/3}$, and it is clear that (as has been emphasized by Hulm) the observed thermal conductivity at low temperatures is considerably greater than the theoretical value even in metals like sodium and copper, to which the theory is most likely to apply. The non-existence in monovalent metals of the theoretically predicted minimum in κ at higher temperatures (Berman and MacDonald 1951, 1952) is evidently a related phenomenon. It should be noted, in this connection, that the theoretical formula for $\kappa(T)/\kappa_\infty$ involves the two parameters T/Θ and D/ζ, and the exact form of the temperature variation depends sensitively on the value which is chosen for D/ζ. The temperature variation of the electrical conductivity, on the other hand, is entirely independent of the value of D/ζ, and the behaviour of the thermal conductivity thus provides a much more searching test of the theory than does that of the electrical conductivity. Further, the value Θ_σ of Θ which has to be used in interpreting the electrical conductivity data (and which should presumably be used also for the thermal conductivity) is usually widely different from the expected value Θ_{L}, that which corresponds to the longitudinal lattice vibrations (Blackman 1951); thus for sodium $\Theta_\sigma=202$, $\Theta_{\text{L}}\simeq 270$, and for copper $\Theta_\sigma=330$, $\Theta_{\text{L}}\simeq 520$. In the present state of the theory, therefore, Θ_σ has to be treated in effect as an arbitrary parameter, whose precise value depends on factors which have been neglected in the derivation of the theoretical formulae. Now the usual relation giving D/ζ in terms of the number of free electrons per atom (Sondheimer 1952) can be expected to hold quantitatively only if the simplifying assumptions of the theory are valid; since they are not valid, it is reasonable to postulate that D/ζ should also be treated as an arbitrary parameter, whose value is of the same order as that suggested by the number of free electrons, but not necessarily equal to it. If, for example, we suppose that the effective value of D/ζ in monovalent metals is about twice the normal value, the agreement between theory and experiment is very much improved, especially if the higher approximations to the theoretical thermal conductivity are taken into account (Sondheimer 1950). (Note here that the minimum in κ is only present if $D/\zeta<1{\cdot}7$.) As we have seen, however, it is not possible, with any reasonable choice of Θ, to fit the experimental data at low temperatures completely with any one value of the parameter D/ζ.

A more drastic procedure, analogous to that used in the analysis of specific heat data, is to regard both Θ and D/ζ as functions of the temperature rather than as constants. It is then possible to adjust the two parameters at any temperature to give the right values of both $\sigma(T)/\sigma_\infty$ and $\kappa(T)/\kappa_\infty$. The values of D/ζ required at low temperatures to obtain agreement with experiment are, however, so widely different from the normal value that the procedure has only formal significance. It seems clear, therefore, that the discrepancies encountered in the case of the thermal conductivity, while not quite so unexpected as has sometimes been suggested, cannot be satisfactorily explained until a more exact theory has become available in which the formula for κ is modified substantially.

I wish to thank Mr. A. H. Wilson, Mr. R. S. Leigh, Dr. R. G. Chambers and Dr. M. Blackman for helpful discussions of these topics.

Department of Mathematics,
Imperial College, London.
2nd May 1952.

E. H. SONDHEIMER.

BERMAN, R., and MACDONALD, D. K. C., 1951, *Proc. Roy. Soc.* A, **209**, 368 ; 1952, *Ibid.*, **211**, 122.
BLACKMAN, M., 1951, *Proc. Phys. Soc.* A, **64**, 681.
HULM, J. K., 1950, *Proc. Roy. Soc.* A, **204**, 98.
MAKINSON, R. E. B., 1938, *Proc. Camb. Phil. Soc.*, **34**, 474.
SONDHEIMER, E. H., 1950, *Proc. Roy. Soc.* A, **203**, 75 ; 1952, *Proc. Phys. Soc.* A, **65**, 561.

The 7 MeV Level of ^{12}C

During a cloud-chamber study of internal pair emission from ^{12}C we have found evidence for the decay by pair emission of an excited state at approximately 7 MeV.

The source consisted of 200 mc of polonium deposited in a thin layer on a 1 cm^2 piece of platinum foil. It was covered by a sheet of beryllium (25 mg/cm^2) and enclosed in an evacuated cell having a thin mica window in front of the beryllium. This window faced a similar one in the wall of a cloud chamber containing air at 60 cm Hg pressure. A magnetic field of 550 gauss was used for momentum determination.

Out of 79 measured pairs 72 had a total energy of $4{\cdot}4 \pm 0{\cdot}5$ MeV, while seven had a total energy of $7{\cdot}0 \pm 0{\cdot}6$ MeV.

We have examined and rejected the possibilities that the tracks of the high energy pairs are those of normal pairs which have undergone considerable multiple scattering, or that they are random associations of a positron with a Compton electron.

It is unlikely that the high energy pairs arise from a 7 MeV γ-ray of ^{12}C or ^{13}C converted in the platinum backing. A number of authors have searched for such a γ-ray from Po–Be sources and a limit to its intensity relative to that of the 4·45 MeV line has been set at 0·5% by Terrell (1950) and at 0·2% by Artemov and Vlasov (1951). An intensity ratio of about 1% was reported by Pringle, Roulston and Standil (1950), but Bell and Jordan (1950) produced evidence for regarding as spurious their own similar observations made by the same method.

Phillips, Cowie and Heydenberg (1951) have observed the emission of electron pairs of total energy 7 MeV from ^{11}B bombarded by protons. They suggest that these might originate in a pair-emitting state of ^{8}Be. The pairs we have observed cannot be produced in this way since the energy available for excitation of ^{8}Be is less than 2·1 MeV.

We therefore conclude that they most probably arise from the decay by pair emission of a 7 MeV excited state of ^{12}C.

If we explain the failure to observe 7 MeV quanta from ^{12}C by assigning to the 7 MeV state the same spin and parity as the ground state (0, even) we must expect a partial decay by radiation to the 4·45 MeV state for which we find, by measurements to be published, spin 2 and even parity.

Observations of the neutron groups from Po–Be by Guier, Bertini and Roberts (1952) give 1 : 8 as the ratio of the population of the 7 MeV state to that of the 4·45 MeV state. Since the ratio of 7 MeV to 4·45 MeV pairs is of the order 1 : 10 and since the pair conversion coefficient for the 4·45 MeV state is of order 10^{-3}, it may be deduced that the probability of pair emission from the 7 MeV state is small compared with that of radiative transition to the 4·45 MeV state.

We should therefore expect the emission from Po–Be of a γ-ray of energy about 2·5 MEV and of approximately one-eighth the intensity of the main 4·45 MEV line. This intensity would probably not have been detected in the experiments already mentioned or in those of Bame and Baggett (1951).

The Clarendon Laboratory,
Oxford.
22nd May 1952.

G. HARRIES.
W. T. DAVIES.

ARTEMOV, K. P., and VLASOV, N. A., 1951, *Dokl. Akad. Nauk, SSSR.*, **77**, 225.
BAME, S. J., and BAGGETT, L. M., 1951, *Phys. Rev.*, **84**, 891.
BELL, P. R., and JORDAN, W. H., 1950, *Phys. Rev.*, **79**, 392.
GUIER, W. H., BERTINI, H. W., and ROBERTS, J. H., 1952, *Phys. Rev.*, **85**, 426.
PHILLIPS, G. C., COWIE, D. B., and HEYDENBERG, N. P., 1951, *Phys. Rev.*, **83**, 1049.
PRINGLE, R. W., ROULSTON, K. I., and STANDIL, S., 1950, *Phys. Rev.*, **78**, 627.
TERRELL, J., 1950, *Phys. Rev.*, **80**, 1076.

An Investigation of the Decay Scheme of ^{153}Sm

Previous work on 47-hour ^{153}Sm is summarized in Nuclear Data (1950). Although there is a considerable measure of agreement on the radiations emitted, two different decay schemes have been proposed by Muehlhause (Nuclear Data 1950) and by Hill and Shepherd (1950). These decay schemes are inconsistent, the total disintegration energies being of the order of 950 kev and 800 kev respectively. The integrated spectrum technique (Bannerman, Lewis and Curran 1951) was therefore employed as a direct means of testing the validity of either scheme. A thin source ($\sim 5\,\mu g/cm^2$) was clamped between the two halves of a 1 cm^3 crystal of NaI(Tl), thus obtaining practically 100% absorption for all radiations. The crystal system was mounted on an EMI 5311 photomultiplier tube and the whole surrounded by 2 in. of lead shielding. The end-point of the integrated spectrum occurred at 800 ± 20 kev, and therefore agrees with the decay scheme of Hill and Shepherd. All features of the low energy region of the spectrum (fig. 1) decayed with the 47-hour half-life of ^{153}Sm. The main peak at 100 kev further confirms the scheme of Hill and Shepherd, but the other features indicate a more complex final solution. Further work on the source can be summarized as follows.

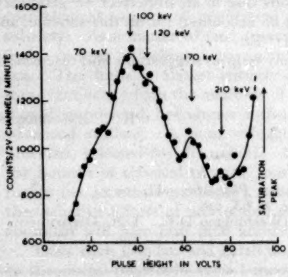


Fig. 1. The ^{153}Sm integrated spectrum.

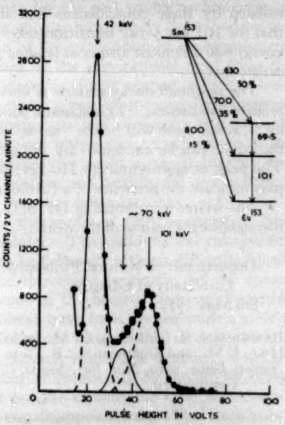


Fig. 2. The γ-ray spectrum from ^{153}Sm.
Inset. The proposed decay scheme of ^{153}Sm.

The γ-ray spectrum. The spectrum observed in a 0·5 cm^3 crystal of NaI(Tl) (fig. 2) showed two lines at 101 ± 2 kev and 42 ± 0·5 kev. This second line is the Eu K x-ray (energy 41·5 kev) arising from internal conversion. No γ-rays were found above an energy of 101 kev, and an upper limit of 0·005% can be given for the intensity of the 610 kev γ-ray. An asymmetry on the low-energy side of the 101 kev peak was shown to be due to the emission of a few unconverted 69·5 kev γ-rays (see fig. 2). The relative intensities of the γ-ray and x-ray lines at 41·5, 69·5 and 101 kev were found to be 70%, ~4% and 26% respectively.

γ–γ coincidence measurements. The spectrum of quanta in coincidence with 101 kev γ-rays consisted of a single x-ray peak, indicating the presence of a highly converted γ-ray in cascade with the 101 kev transition. It was calculated from the geometry that one in every three 101 kev γ-quanta was accompanied by an x-ray in coincidence. Thus about 70% of the total K x-ray intensity is due to internal conversion of the 101 kev γ-ray, the remaining 30% arising from the 69·5 kev transition. Allowing for the fluorescence yield in the K shell of Eu and for L conversion (K/L ~5/1, Hill and Shepherd 1950), a value of the order of 3 is then obtained for the internal conversion coefficient of the 101 kev γ-ray. No separate peak due to 69·5 kev γ-rays was observed in the coincidence spectrum, but an upper limit of one-tenth the K x-ray peak can be given for its intensity. The internal conversion coefficient is therefore greater than 10.

The β-ray spectrum. A thin source (~5 μg/cm^2) and a 0·5 cm^3 crystal of NaI(Tl) were used to examine the β-ray spectrum. The solid angle was kept small to reduce coincidences from the source, and the end-point was confirmed at 800 ± 20 kev. The highest energy β-ray therefore goes directly to the ground state of ^{153}Eu.

β–γ coincidence measurements. Under suitable conditions of geometry, coincidences were observed between β-particles and 101 kev γ-rays. The β-spectrum obtained when isolated by these coincidences had an end-point at 700 ± 20 kev. This indicates clearly that the 101 kev γ-ray transition takes place to the ground state of ^{153}Eu. A third β-ray of energy 630 kev must also exist leading to the 69·5 kev transition, but no attempt was made to isolate this.

The resultant decay scheme is illustrated in fig. 2, inset, with approximate values for the relative intensities. This scheme accounts directly for the peaks at 100 kev and 170 kev in fig. 1, since these will be the ‘ zeros ’ of the two displaced β-spectra. The other features of the curve can be explained by the escape of radiations due to an imperfect 4π geometry. The peak at approximately 210 kev, however, cannot be accounted for on this scheme, and may indicate the presence of a further excited level in ^{153}Eu.

The writer is indebted to Dr. S. C. Curran for many helpful suggestions and discussions throughout the course of the work.

Department of Natural Philsophy,
University of Glasgow.
8th May 1952.

R. C. Bannerman.

Bannerman, R. C., Lewis, G. M., and Curran, S. C., 1951, *Phil. Mag.*, **42**, 1097.
Hill, J. M., and Shepherd, L. R., 1950, *Proc. Phys. Soc.* A, **63**, 126.
Nuclear Data, 1950, *Nat. Bur. Stand. Circular No.* 499 (Washington D.C.: U.S. Department of Commerce).

REVIEWS OF BOOKS

Integral Transforms in Mathematical Physics, by C. J. Tranter. (Monographs on Physical Subjects). Pp. ix + 118. (London: Methuen, 1951.) 6*s*.

The principal aim of this book is to outline the use of integral transforms in the solution of partial differential equations. The solution of an ordinary differential equation can often be reduced to that of an algebraic equation by the use of an integral transform. In a similar manner a partial differential equation in *n* independent variables can be reduced to one containing $(n-1)$ variables. The method facilitates the solution of such equations with given boundary and initial conditions.

The following integral transforms are treated: Laplace, Fourier sine and cosine, complex Fourier, Hankel and Mellin. The corresponding inversion formulae are obtained formally. Separate chapters are then devoted to applications of these transforms, each being illustrated by the solution of problems—many of which are taken from recent papers by the author. The student of electric circuits will perhaps be disappointed to find that practically every example relates either to heat conduction or to elasticity.

Of special interest are the two chapters dealing respectively with the numerical evaluation of 'inversion' integrals, and finite Fourier and Legendre transforms. The last chapter shows how a partial differential equation in three variables may be reduced to one in two, which latter can then be solved by relaxation methods.

Those with a limited background of mathematics will need to acquaint themselves with the elements of analysis and with the evaluation of integrals using contour integration. Given this equipment the book will be welcomed as a compact and workmanlike exposition of a subject which has come into great prominence in many fields. H. H. HOPKINS.

The Classical Theory of Fields, by L. LANDAU and E. LIFSHITZ. 2nd Edition. Pp. ix + 354. (Cambridge, Mass.: Addison-Wesley, 1951.) $7.50.

The well-known vigour and originality of thought of the senior author of this book, and the mathematical virtuosity of his eminent collaborator, confer a special interest on this product of their didactic activity. The present volume gives us only a fragmentary view of their conception of a course of theoretical physics. Judging from the excellence of this part, one would wish to have the whole work as carefully translated and neatly produced. The plan of the course must be quite unconventional, for the part under review presents a rather unfamiliar assemblage of topics, which are only approximately described by the title. Essentially, what we are given is a discussion of the production of electromagnetic fields *in vacuo* by charged particles, and a treatment of the gravitational field from the point of view of general relativity. However, the electromagnetic section, which is built up from the start on the theory of relativity, also includes a brief, but masterly, exposition of geometrical optics and diffraction. It is gratifying to find a simple and accurate discussion of the nature and production of gravitational waves and an exposition of the cosmological aspects of gravitation theory distinguished by an exceptional lucidity and sobriety. As a matter of fact, one may pick up at random any section and one is sure to find a concise and elegant treatment, in which the essential points are admirably brought out. The book is almost entirely self-contained; the reader is only assumed to have some familiarity with the methods of analytical mechanics. The mathematical calculations, though condensed, are always sufficiently explicit to be followed without difficulty by the advanced student. A most valuable feature is the refreshing originality of the special problems selected for discussion. Although the scope of the book is kept rigidly within the bounds of classical physics, most of these problems have been chosen with a view to future use in quantum theory; others have by themselves considerable interest, such as the radiative effects of accelerated particles in the extreme relativistic domain. Even the specialist will here find little known results of recent investigations by Soviet physicists.

Does then Landau and Lifshitz's course at last provide us with the ideal introduction to theoretical physics? Here I must, alas! express my disappointment. The stern beauty of a smooth flow of rigorous mathematical deductions is bought at a heavy price. It actually amounts to putting the natural development of the subject upside down. I know from our discussions in the good old days that Landau's 'violon d'Ingres' has always been his belief that one could encompass the whole of physics in a monumental 'principle of least action'. So far as I can judge, this is the leading idea which he has unflinchingly carried out in this course, and we can see the result. A few specific examples will show better than much philosophical discourse the kind of incongruities which this attitude entails. To write down the principle of least action governing the relativistic motion of a particle we have just to look for a suitable Lorentz invariant, and there is no other than the interval. This sounds fine, but if we ask for the physical basis of the requirement of Lorentz invariance we must work our way back to page 1, only to strand on the statement: " Experiment shows that . . . all the laws of nature are identical in all inertial systems of reference." The introduction of the electromagnetic field is much worse still. The additional term in the Lagrangian, containing the potential, is simply thrust upon the reader with the comforting words: "It is found to have the form . . ." And on this basis the first pair of Maxwell equations is 'derived'! It need not be emphasized how misleading and didactically unsound such a procedure is.

The debate is not new, of course. The idea of treating mechanics as a 'deductive' science goes as far back as Lagrange. Commenting on the *Mécanique analytique*, that profound mathematician Poinsot shrewdly observed that if one tried to introduce into that great book a 'proof' of the 'principle of virtual velocities', "l'ouvrage se trouverait fait deux fois : je veux dire que cette démonstration comprendrait déjà toute la mécanique". It seems to me that there is no escape from this situation. One might say, with a grain of salt, that theoretical physics should be learned twice over. The first stage should proceed inductively and historically, so as to retrace the dialectical unfolding of the subject. This stage would indeed culminate in the establishment of great universal principles; it would show these principles in their true light, as a condensation of experience in abstract, idealized form, with all the inherent limitations of their range of validity. Only then would the ground be safe for a second survey in the reverse direction. The principles could then be used as a basis for a mainly deductive analysis of more delicate problems, in the treatment of which they would demonstrate their real fruitfulness. For this return journey the advanced student could find no better guide than our authors. But for his first introduction to the true spirit of theoretical physics he will still have to rely on the old masters, Abraham and Sommerfeld, who (albeit unwittingly) display that fine sense for dialectics which is so strangely lacking even in the best productions of our Russian friends.

L. ROSENFELD.

Dictionary of Mathematical Sciences: Vol. 1, by L. HERLAND. Pp. 235. (London: Hoffner, 1951.) 24*s*.

This work claims to be the first modern bilingual dictionary of mathematics. The vocabulary is mainly that associated with pure mathematics and mathematical physics; but, in addition, a very large number of entries seem to be commercial terms and phrases. Moreover, many terms from such fields as physics and astronomy are included.

There are some surprising entries to be found within the pages of this volume. It seems unnecessary to explain that 'chi (griechischer Buchstabe)' has the meaning 'chi'—nor, indeed, to give corresponding 'meanings' of the other letters in the Greek alphabet. Again, the now rarely used, 'q.e.d.' is given under 'quod erat demonstrandum (cf. Q.E.D.)' and again under 'q.e.d. (abbr.) quod . . . etc. (Lat.) (was zu bewiesen war, which was to be demonstrated) (abbr.) Q.E.D.'. This seems to be a sheer waste of space.

On the other hand a careful check of about fifty technical words used in mathematics revealed only one omission—namely 'Aufpunkt'. A general browsing revealed occasionally odd English (?) words; e.g. catacaustic.

The typography is extremely clear, and the general lay-out of the book is commendable. Against each word is an indication of what part of speech it is, and the genders of nouns are given—but, unfortunately, not their plural forms.

There is no doubt that most libraries and many individuals will find it necessary to possess both this and the subsequent volume.

H. H. HOPKINS.

Atomic Scientists' News, Vol. 1, No. 1. (London: Taylor & Francis, September 1951.) Published bi-monthly, 6*s*. per issue.

This is the first issue of a new series of the journal of the Atomic Scientists' Association, intended for the general public as well as for the members of the Association.

Even if the articles were expressions only of personal opinion their general tenor would inevitably be taken by the public as the 'policy' of the Association, and it is clear from this issue, and in particular from the account of the annual general meeting, that it is aware of the dangers this would involve. It can offend nobody by having no policy at all on the development of nuclear energy, or it can arouse enthusiasm but only among the few by presenting a forthright policy with probable political implications. The journal is at pains to avoid either extreme by adopting a policy primarily of scientific accuracy. It is an open question whether this will always keep the journal free from the charge of being political, but it is a policy to which no honest scientist can object. That it can be used with effect is shown by the devastating analysis of an article on *Russian Atomic Power* in *World Review.*

The longest article is a review of recent progress in nuclear physics and engineering and related political events, and there is a most useful comprehensive account by Sir John Cockcroft of the long-term possibilities of nuclear power. The reader may care to guess what is concealed beneath Professor Peierls' title *Bathwater and the Baby.*

It is a well-balanced issue although the scientific terminology must prove rather difficult for the ' general ' public. If it continues in this manner the journal should be very useful in keeping the average scientist well informed about the physical facts and the social implications of the most dangerous branch of his activities. N. C. B.

Advances in Physics : A Quarterly Supplement of the 'Philosophical Magazine', Vol. 1, *No.* 1, *January* 1952. Editor : Professor N. F. MOTT, F.R.S. Pp. 109. (London : Taylor and Francis.) 15*s.*

Advances in Physics is a supplement to the *Philosophical Magazine* which will appear quarterly, from January of this year. The Editor, in his introductory remarks about the new journal, describes its purpose as follows : " The aim of the journal is to publish specialist articles, of admittedly ephemeral value, in rather narrow fields, and to publish them quickly ".

It is natural to compare the journal with the Physical Society's annual *Reports on Progress in Physics.* Some differences are already clear. The intended practice of publishing small quarterly numbers, each containing about three articles linked to some extent in their subject matter, rather than large annual volumes covering diverse branches of physics, will open the way towards giving each number a specialist appeal rather than a wide interest. The effect of this, together with the policy of inviting 'stop press' accounts of subjects at present in a state of flux, should be that the new journal will complement the *Reports.* One imagines that people will turn to the *Reports* to see how the frontiers of physics are advancing as a whole, and to the *Advances* to join in forays and thrusts that are now being made in particular fields.

The three articles in this opening number deal with problems of crystals. Dr. Sondheimer writes on the mean free paths of electrons in metals, Professor Seitz on the generation of vacancies by moving dislocations, and Dr. Frank on crystal growth and dislocations.

Dr. Sondheimer gives a most interesting account of recent work on methods of finding the mean free path of electrons in metals. The idea in all cases is to measure electrical resistance under such circumstances that this path is about equal to some other characteristic length in a metal. The simplest case occurs with thin films at low temperatures, where the mean free path may equal the thickness of the metal. Another case is a magneto-resistance effect, where the important length is the radius of a free electron orbit in a magnetic field. Lastly, there is the anomalous skin effect, which involves the depth to which a high frequency electric field penetrates a metal.

Professor Seitz accepts the Editor's hint by concentrating intentionally on ideas that are still mainly speculative. His main point is that vacant lattice sites can be generated from moving dislocations. The consideration of special processes, such as intersecting screw dislocations cutting through one another, shows that this idea is very reasonable, and several experiments exist which go some way towards confirming it. In his final pages Professor Seitz speculates in a most interesting and provocative way about the origins of work hardening.

Dr. Frank in his article describes a delightful chapter of post-war physics, his own dislocation theory of crystal growth and its dramatic confirmation by the work of Griffin, Dawson and others. It seems hardly necessary even to read Dr. Frank's closely reasoned argument, for the beautiful photographs speak so eloquently in support of the main idea of his theory, which is that growth is catalysed at the points where dislocation lines emerge on the surface of the crystal.

The Editor is to be congratulated on having made such a good start to his new journal; it will whet the reader's appetite for future numbers.

A. H. COTTRELL.

Fourier Transforms, by IAN N. SNEDDON. Pp. xii + 542. (London: McGraw-Hill, 1951.) 85*s*.

The subject matter of this book is the practical solution of the boundary value problems of physics by means of integral transforms. Despite its title, it deals not only with those transforms in which the kernel is a trigonometrical function but also gives a detailed treatment of the Hankel transform and brief accounts of the Mellin and Laplace transforms. The first ninety pages are used to develop the necessary formulae for the transforms and their inversions; the rigour of the treatment is sufficient to meet most of the requirements of mathematical physics. The remaining four-fifths of the book show how the transform method has been applied to the solution of problems in many different parts of physics. Successive chapters are devoted to vibrations, heat conduction, neutron diffusion, hydrodynamics, atomic and nuclear physics, and stress distributions. Each of these chapters consists of a collection of examples, mostly taken from recent research applications of the transform technique. The examples have been chosen for their practical importance, rather than as illustrations of the method, with the result that the calculations are often extremely complicated. Many of them, particularly in the field of elasticity, are the original work of the author.

Several of the examples show the value of using the less well-known methods of finite transforms and dual integral transforms. Relatively little space is given to the Laplace transform, but the author shows that it is sometimes advantageous to use other transforms in place of the Laplace one. It would have been useful if the author had indicated how it is possible to decide on the most suitable transform for any particular problem. One of the appendices consists of a short list of some of the commoner transform pairs. A more complete list would have been welcomed since, although other lists have been published, they are not always easily accessible.

The greatest merit of this book is the vitality which it gains from dealing with problems of real practical interest. Inevitably only a small proportion of these problems will be of interest to any one physicist, but they may well suggest other applications of the transform methods. For this purpose the book might have been improved if there had been rather fewer examples in the text but more discussion of the conditions under which the transform methods are likely to succeed.

J. C. B.

Photoelectric Tubes, by A. SOMMER. 2nd Edition, revised. Pp. viii + 118. (London: Methuen, 1951.) 8*s*. 6*d*.

This volume represents the second edition of *Photoelectric Cells* previously published in this series by the same author. The new title gives a more accurate description of the subjects under discussion which include all types of electronic tubes based on photoelectric emission and exclude photovoltaic and photoconductive cells which depend on barrier layer mechanisms. A new chapter has been added (contributed by Mr. S. Rodda) which deals with some of the fundamentals of photoelectric emission processes. This addition should certainly prove helpful to the type of reader for whom the book is intended, though there are some parts which may prove a little difficult, e.g. those dealing with the behaviour of electrons in surface layers and with photo-ionization. Other topics discussed in the volume include a description of various types of photo-cathodes, problems concerned with spectral sensitivity and the matching of phototubes to particular light sources, the operating mechanisms of gas-filled photocells and the properties of multiplier photocells. There is also a brief survey of applications in photometry, sound film reproduction and television, and a useful bibliography gives the sources from which more detailed information can be obtained. The book is generally well written and will commend itself particularly to research students and to others who need an introduction to this important subject.

H. K. H.

Variations et origine du rayonnement cosmique, by A. Dauvillier. Pp. 83. (Paris : Éditions de la Revue d'Optique Théorique et Instrumentale, 1951.) No price given.

The study of the variations of the cosmic-ray intensity with time has already yielded much valuable information about the origin of the radiation, and it is appropriate, therefore, that these two aspects of cosmic-ray physics should be treated together in this book. In the first chapter there are sections dealing with the periodic and non-periodic intensity changes. These include the solar and sidereal daily variations, the lunar variation, the seasonal variation and an account of the intensity changes associated with magnetic storms and solar activity. The second chapter reviews some of the theories and speculations about the origin of the radiation which have been advanced during the past thirty years or so. These include the annihilation hypothesis of Millikan, the cosmological theories of Lemaître and Milne, and the more recent theories which ascribe the origin of the radiation either to stellar magnetic fields or to weak magnetic fields in interstellar space. These topics have received very little attention from previous authors of books on cosmic-ray physics, and this book will be of value to all who are interested in the geophysical and astrophysical aspects of the subject.

H. ELLIOT.

Combustion, Flames and Explosions of Gases, by B. Lewis and G. von Elbe. Pp. xix + 795. (New York: Academic Press, 1951.) $13.50.

.A
5
2

In the past, progress in knowledge of combustion phenomena and the development of chemistry have gone closely together, from the days of the phlogiston theory to the important developments in chemical kinetics and the chain propagation theory around 1930. The more recent advances in combustion seem, however, to be more closely linked with various branches of physics. This trend is reflected in the present book, in which a bare quarter is devoted to the purely chemical processes, and the remainder largely to such subjects as wave propagation, turbulence, heat transfer, thermodynamics, diffusion and minimum ignition energy.

While the book retains the same title as the first edition in 1938, it is in fact almost entirely new, nearly all of it being rewritten, incidentally from a much more mature viewpoint, and it is over twice the original size. The object is still to provide the chemist, physicist and engineer with the scientific basis for understanding combustion phenomena. It will undoubtedly become and remain for a long time the standard reference book on the subject.

The book is divided into four main parts, headed ' Chemistry and kinetics of the reactions between fuel gases and oxygen ', ' Flame propagation ', ' State of the burned gas ' and ' Problems in technical combustion processes '. There are also some valuable appendices giving numerical and graphical data for thermochemical calculations, limits of inflammability and flame temperature.

The first part discusses both the general theory of chemical kinetics and the many individual reactions occurring in the combustion of hydrogen, carbon monoxide and hydrocarbons. This is an actively developing field, but there now exists a considerable area of agreement among workers and the more controversial aspects are not unduly stressed. Indeed the new chemical mechanisms put forward here may help to stabilize knowledge of the subject. The second part, on flame propagation, occupies about half the book; this deals with the propagation of the combustion wave in non-turbulent and turbulent mixtures and detonations, and is especially valuable for its discussion of the many aspects of flame stability and for the integration of the concepts of minimum ignition energy with results for quenching distances, spark ignition and burning velocity. The whole treatment here is quantitative and precise. While the emphasis is perhaps on the basic scientific theory, the results are illustrated all through by excellent flame photographs of various types, many numerical data and many graphs and figures. On the experimental side the principles of flame photography by direct, shadow, schlieren and interferometric methods are described, and also the determination of pressure, and of flow pattern by particle track photography. The sections on measurement of burning velocity and of flame temperature are perhaps a little brief and rather lost. There are some excellent photographs and an interesting account of the cellular structure of some flames, and of the so-called polyhedral flames; the importance of the selective rates of diffusion of the gas constituents in causing these effects is stressed.

On the whole this is a big task very well accomplished. The authors, in addition to their own great experience, have been able to call on the co-operation of several of the staff at the U.S. Bureau of Mines, and the whole book is a thoroughly authoritative treatment of the subject. There is obviously some difficulty in arranging a complex subject like combustion; some of the sections are very long, that on cool flames and two-stage ignition running for 33 pages and that on ignition by electric sparks for 35 pages without any sub-headings at all. Particularly valuable features are the numerous schlieren photographs of flame structure, and diagrams of flow patterns. The photographs and illustrations are a great asset in a book of this type. A. G. G.

Progress in Cosmic Ray Physics, Ed. J. G. WILSON. Pp. xvi+557. (Amsterdam: North Holland Publishing Co., 1952.) 86*s*.

The output of papers on the cosmic radiation is now so large that any attempt to assist the assimilation of the available data is certain to be welcomed. The present volume consists of eight separate articles, which are written by different authors on branches of the subject in which they are authorities. All the articles are primarily intended for those engaged on cosmic-ray research, but some of them may also be useful to those working on other subjects. The stress is always on the physical results rather than on the experimental methods.

The four most successful reviews are those dealing with high energy nuclear disintegrations, the nature of the primary cosmic radiation, observations underground and time variations. In each case the subject matter is sufficiently limited to enable that a fairly complete survey can be given. They are all most valuable in showing clearly both the extent of the knowledge which has been acquired and the problems which still require further investigation. The only theoretical review gives an account of the various types of fundamental particle and the possible couplings between them. It is a remarkable achievement that an article on theoretical physics should contain hardly any mathematics; even so, and despite the obvious care of the author, it is still by no means easy for an experimentalist to follow all the arguments. An article on the decay processes of heavy mesons suffers from the paucity of results available—it reads more like an original paper than a review article. A chapter which deals with recent data on geomagnetic effects contains a useful account of some experiments but does not review all the results which give information on these problems. An article on the equilibrium of the cosmic-ray beam in the atmosphere is the only one which overlaps with any of the others. The scope of this article is very wide; possibly in consequence of this, the data have not always been considered sufficiently critically.

It will be seen that although this volume is not intended to be a complete survey, it does give an account of most of the more rapidly developing parts of the subject. The general production of the book is very good. Each section has its own list of references, but there is also a general author index—almost a bibliography of cosmic rays—and a subject index. The reproduction of the photographs is so good that the varying quality of the originals is clearly revealed. There are very few errors or inconsistencies between the various authors. The editor and publishers are especially to be congratulated on producing such a complicated work in less than six months. The high cost is the only consideration which may deter some cosmic-ray physicists from acquiring this excellent volume. J. C. B.

Control and Removal of Radioactive Contamination in Laboratories, U.S. Department of Commerce, National Bureau of Standards. Pp. iv + 24. (Washington : Government Printing Office, 1951.) 15*c*.

X-ray Calibration of Radiation Survey Meters, Pocket Chambers and Dosimeters, by FRANK H. DAY, National Bureau of Standards Circular 507. Pp. 11. (Washington D. C. : U. S. Department of Commerce, 1951.) 15*c*.

Energy and Angle Distribution of the Photoprotons from Deuterium, by MARTIN WIENER, National Bureau of Standards Circular 515. Pp. ii + 13. (Washington D.C. : U.S. Department of Commerce, 1951.) 15*c*.

Nuclear Data, Supplement 2 (July 1950 to January 1951) to National Bureau of Standards Circular 499. Pp. 63. (Washington, D.C. : U.S. Department of Commerce, 1952.) Price, $4.25, includes Circular 499 and three supplements, the third of which is scheduled for publication in 1952.

CONTENTS FOR SECTION B

PAGE
Dr. F. C. CHAMPION. Electrical Counting Properties of Diamonds 465
Mrs. K. STRATTON and Dr. F. C. CHAMPION. Electrical Counting Response of Two Large Diamonds under Beta-Irradiation 473
Prof. A. L. REIMANN and Mr. J. V. SULLIVAN. Rectification Phenomena Exhibited by Natural and Sulphurized Galena 480
Mr. A. N. GENT and Mr. R. S. RIVLIN. Experiments on the Mechanics of Rubber : II—The Torsion, Inflation and Extension of a Tube 487
Mr. P. GROOTENHUIS, Dr. R. W. POWELL and Mr. R. P. TYE. Thermal and Electrical Conductivity of Porous Metals made by Powder Metallurgy Methods 502
Mr. R. S. BARNES. Effects associated with the Flow of Vacancies in Intermetallic Diffusion 512
Mr. A. R. VERMA. New Observations of Crystal Overgrowth on Silicon Carbide . 525
Mr. F. R. L. SCHOENING, Dr. J. N. VAN NIEKERK and Dr. R. A. W. HAUL. Influence of the Apparatus Function on Crystallite Size Determinations with Geiger Counter Spectrometers 528
The late Mr. H. LUND and Mr. L. WARD. The Spectral Emissivities of Iron, Nickel and Cobalt 535
Dr. K. E. SPELLS. A Study of Circulation Patterns within Liquid Drops moving through a Liquid 541
Mr. E. W. B. GILL and Mr. G. F. ALFREY. The Electrification of Liquid Drops . 546
Letters to the Editor :
    Prof. P. SELÉNYI. The Influence of Mercury Vapour on Selenium Rectifiers and Selenium Photoelements 552
    Mr. H. A. GEBBIE and Mr. D. G. KIELY. The Dielectric Constant and Loss of Amorphous Selenium at a Wavelength of 3 cm 553
    Dr. R. J. WEISS. Extinction Effects in Powders 553
Corrigenda 555
Reviews of Books 555
Contents for Section A 558
Abstracts for Section A 559

.A
5
2

ABSTRACTS FOR SECTION B

Electrical Counting Properties of Diamonds, by F. C. CHAMPION.

ABSTRACT. Examination was made of over 200 diamonds, most of which were of gem quality and many of which weighed only a few milligrams. About 25 of them showed good conduction properties when submitted to ionizing radiations. For depths of crystal penetration which exceeded a certain amount, all the counting specimens responded to both α-particles and β-particles with a response which was at least qualitatively dependent on the energies of the particles. In some cases, however, a marked drop in the counting properties was easily demonstrated with α-particles under conditions of feeble penetration. Even at maximum penetration 5-MEV α-particles produced oscillograph pulses in such crystals which were only about three times as large as those from 1-MEV β-particles. Strong correlation was established between the wavelength of the continuous ultra-violet transmission limit and the magnitude of the electrical counting response of the specimen.

Electrical Counting Response of Two Large Diamonds under Beta-Irradiation, by K. STRATTON and F. C. CHAMPION.

ABSTRACT. Two large diamonds S_1 and S_5, both of good optical quality, were subjected to β-irradiation and the electrical conduction responses were studied under a variety of conditions. With S_1 special attention was paid to the formation and removal of space charge. There is evidence that the mechanisms of space charge removal by β-irradiation and by illumination respectively are not quite the same. The efficiency of this diamond as a β-particle counter was not appreciably different from that of a standard Geiger counter.

Small volume elements of S_1 and S_5 were examined individually for β-counting efficiency. While S_1 did not show a variation greater than 20%, variations as large as ten to one were found in different parts of S_5. The results were not inconsistent with the existence of a correlation between the ultra-violet transmission and the counting efficiency of any selected volume element.

The β-response of S_1 was more than three times the best response from S_5 under comparable conditions, while the ultra-violet transmission limits were 2250 and 2850 Å respectively.

Rectification Phenomena Exhibited by Natural and Sulphurized Galena, by A. L. REIMANN and J. V. SULLIVAN.

ABSTRACT. Specimens of n-type galena were sulphurized, either by a wet chemical process or by exposure to sulphur vapour at various temperatures, and the resulting changes in rectification characteristics were observed, both with d.c. instruments and at 3 000 Mc/s, using various cats-whiskers. According to treatment given, either an improved n-type rectification was obtained or the polarity was changed to p-type. The results are briefly discussed in the light of present-day rectification theory.

Experiments on the Mechanics of Rubber : II—The Torsion, Inflation and Extension of a Tube, by A. N. GENT and R. S. RIVLIN.

ABSTRACT. The formulae derived by Rivlin (1949) for the forces necessary to produce simple extension, radial inflation and torsion in a tube of incompressible highly elastic material, which is isotropic in its undeformed state, are specialized to the case when the amount of torsion is small. The resulting formulae are compared with measurements on tubes of vulcanized natural rubber and conclusions as to the form of the stored-energy function are drawn, in general agreement with those reached from earlier experiments. The experiments indicate that hysteresis between the loading and unloading curves for the vulcanizate is associated with certain terms in the expression for its stored-energy function and, presumably, with the mechanism which accounts for the presence of these terms.

Thermal and Electrical Conductivity of Porous Metals made by Powder Metallurgy Methods, by P. Grootenhuis, R. W. Powell and R. P. Tye.

ABSTRACT. Measurements are reported of the thermal conductivity and electrical resistivity over the range of 20° c to 200° c on several specimens of a sintered porous bronze material (89% Cu, 11% Sn). The constituent powder particles were substantially spherical in shape and the specimens studied covered particle diameters ranging from 0·00133 to 0·040 cm and had densities from 5·27 to 7·01 g/cm^3. The modified Lorenz relationship between the thermal conductivity K the electrical resistivity ρ and the absolute temperature T was found to apply, the experimental results satisfying the straight line $K=0{\cdot}58\times10^{-8}T/\rho+0{\cdot}005$. Existing data for solid bronzes of similar composition also conformed to this line. It is shown that, to a first approximation, the conductivities are independent of the particle size of the powder from which the specimen has been made, but can be correlated to a density basis, the values falling along a straight line drawn from the point for the solid material to cut the abscissa at a density corresponding to the maximum porosity which can be attained on packing equal sized spheres. This method of correlation is also confirmed by data from the literature for the electrical conductivity of copper, nickel and iron. Suggestions are made for estimating the thermal conductivities of similar porous materials.

Effects Associated with the Flow of Vacancies in Intermetallic Diffusion, by R. S. Barnes.

ABSTRACT. Experiments on the interdiffusion of copper–alpha-brass and copper–nickel couples and sandwiches, have shown that there is an increase of volume in the diffusion zone as a result of diffusion. This increase in volume is associated with the formation, in the diffusion zone, of voids with crystallographic faces. These voids appear near to the original interface and on that side from which there is a net loss of atoms, or a net gain of vacancies, as shown by the movement of inert markers. Grain boundaries influence the movement of the markers in regions where there has been preferential grain-boundary diffusion. A close micrographical examination of the diffusion zone shows it to have an unusual structure, various observations pointing to the presence of strain, and x-ray back reflection photographs of a copper–nickel couple show that the diffusion zone is polygonized. Subsidiary experiments suggest how strain in the diffusion zone arises.

A vacancy diffusion mechanism where there is a preferential flow of vacancies in the one direction, is used to explain the above phenomena. This flow will produce strain in the diffusion zone if vacancies are generated on one side of the original interface and collapse on the other. Mechanisms are discussed for the generation and condensation, within the diffusion zone, of the large numbers of vacancies necessary for the process. To explain the volume increase and formation of voids it is postulated that not all the vacancies on the one side of the interface are eliminated by the lattice collapsing, but that some vacancies coalesce to form microscopic voids which enlarge by assimilating further vacancies.

New Observations of Crystal Overgrowth on Silicon Carbide, by A. R. Verma.

ABSTRACT. On the faces of some silicon carbide crystals showing ‘ growth spirals ’ superimposed oriented overgrowths can sometimes be observed. These oriented overgrowths which took place during the process of manufacture consist of stepped triangular pyramids with craters at their peaks, the pyramids having heights up to a few wavelengths of light, and built sometimes on platforms of similar heights. These conclusions have been arrived at by the application of the techniques of multiple-beam interferometry and light-profile microscopy. In addition to the pyramids some trigonal, hexagonal and their compound figures have also been observed, which by the application of multiple-beam interference fringes are shown to be molecular oriented layers. These oriented molecular layers, the existence of which is revealed from the observation of the above overgrowths on silicon carbide crystals, may be silica deposit or possibly the growth of silicon carbide on itself.

Influence of the Apparatus Function on Crystallite Size Determinations with Geiger Counter Spectrometers, by F. R. L. Schoening, J. N. van Niekerk and R. A. W. Haul.

ABSTRACT. A brief survey of the various methods used for determining crystallite sizes from x-ray diffraction line broadening is given. It is shown that the diffraction profiles determined experimentally with a Geiger counter x-ray spectrometer can be represented by functions of the type $y=c/(1+k^2x^2)^2$. Using these functions a correction curve for instrumental influence on line breadth measurements is derived by solving the integral equation $B(x)=\int_{-\infty}^{+\infty} b(v)\beta(x-v)dv$ by means of the Fourier transform method. The apparatus function $b(x)$ is determined by using a standard substance. Experiments are described which illustrate the effect of sample preparation, absorption, etc. on measurements of line breadth.

The Spectral Emissivities of Iron, Nickel and Cobalt, by the late H. Lund and L. Ward.

ABSTRACT. The spectral emissivities of iron, nickel and cobalt have been measured by the direct comparison of surface and black body radiation at temperatures between 1 000° c and 1 300° c, over the wavelength range $1{\cdot}0\,\mu$ to $2{\cdot}6\,\mu$, using a lead sulphide cell and amplifier as the detecting unit. Values of the temperature coefficients of emissivity referred to 1 000° c indicate an X point for iron at $1{\cdot}6\,\mu$, but none for nickel in this spectral range. The results for cobalt show an anomaly at a temperature close to the Curie point.

A Study of Circulation Patterns within Liquid Drops moving through a Liquid, by K. E. Spells.

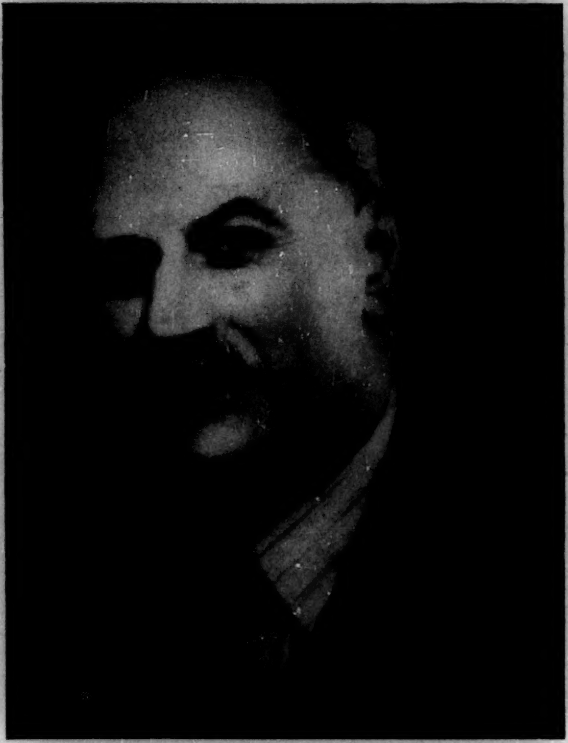
ABSTRACT. The technique used to reveal the circulation patterns was developed by Hagerty, and depends on the striae which appear in glycerine and glycerine–water mixtures when sheared.

Instantaneous flash photographs were taken of circulation patterns in drops of glycerine falling slowly (about 1 cm/sec) in castor oil, and in drops of various glycerine–water mixtures falling at about 10 cm/sec in heavy white oil (viscosity 0·37 poise at 15° c). In the former case the drops were spherical and the circulation pattern agreed reasonably well with that predicted theoretically by Hadamard; in the latter case the drops were no longer spherical and the centre of circulation was displaced below the equatorial plane.

The Electrification of Liquid Drops, by E. W. B. Gill and G. F. Alfrey.

ABSTRACT. Charges on drops breaking up in electric fields were investigated and it was found they could all be explained by the ordinary laws of electrostatic induction, with allowance for contact potential differences. Electrification due to splashing was also studied and again the effects could be explained by contact potentials alone so that theories put forward of double layers to explain previous experiments were unnecessary. A warning is given of the necessity of making due allowance for possible electrification by splashing in interpreting experimental results and a critical examination of an experiment of Zeleny is reported.

A few experiments of splashing water drops off ice gave results which may have a bearing on the production of electricity in thunderstorms.



PROFESSOR L. F. BATES, D.Sc., Ph.D., F.R.S.
President of the Physical Society 1950–52.

(The Presidential Address is published in the August issue of the *Proceedings*, A, page 577.)